

## BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

Paper No. 93

Application Number: 08/303,561

Filing Date: September 09, 1994

Applicants: Johannes G. Bednorz et al.

Examiner M. Kopec

Daniel P. Morris For Appellant

## SUBSTITUTE SUPPLEMENTAL APPELLANT'S REPLY TO THE EXAMINER'S ANSWER TEXT ONLY

In the appellants' reply reference was made to the article "Synthesis of Cuprate Superconductors" by Rao et al., IOP Publishing Ltd. 1993. A copy of this article is in Attachment C to the reply brief. This article lists in Table 1 the properties of 29 cuperate superconductors made according to appellants teaching. Twelve (#'s 1, 8-13, 16, 17, 20, 21, 27 and 28) of those listed do not come within the scope of the claims allowed by the examiner. Only three of the 29 have a Tc < 26°K. Those twelve do not contain one or more of a rare earth, a group IIIB element or an alkaline earth element. It is thus clear that broader claims than allowed in the answer should be allowed since it is clear that the allowed claims can be avoided following appellants' teaching without undue experimentation.

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Docket YO987-074BY

The article of Rao et al. in the first sentence of the introduction citing appellants' article - which is incorporated by reference in their application - acknowledges that appellants' initiated the field of high Tc superconductivity. Appellants further note that the Rao article acknowledges that "a large variety of oxides" are prepared by the general principles of ceramic science and that appellants discovered that metal oxides are high Tc superconductors.

Citing reference 5 therein - the book "New Directions in Solid State Chemistry", Rao et al. 1989 (Cambridge; Cambridge University Press) for which there is a 1986 edition which predates appellants filing date Rao (See Attachment B) - Rao et al. states:

Several methods of synthesis have been employed for preparing cuprates, with the objective of obtaining pure monophasic products with good superconducting characteristics [3, 4]. The most common method of synthesis of cuprate superconductors is the traditional ceramic method which has been employed for the preparation of a large variety of oxide materials [5]. Although the ceramic method has yielded many of the cuprates with satisfactory characteristics, different synthetic strategies have become necessary in order to control factors such as the cation composition, oxygen stoichiometry, cation oxidation states and carrier concentration. Specifically noteworthy amongst these methods are

chemical or solution routes which permit better mixing of the constituent cations in order to reduce the diffusion distance in the solid state [5, 6]. Such methods include coprecipitation, use of precursors, the sol-gel method and the use of alkali fluxes. The combustion method or self-propagating high-temperature synthesis (SHS) has also been employed.

Reference 5 is another example of a reference to the general principles of ceramic science incorporated into appellants' teaching. The Rao et al. article states that the 29 materials reported on in the article and listed in Table 1 are fabricated using the general principles of ceramic science. Moreover, the Rao article states that these materials are fabricated by what the Rao article calls the "ceramic method" which is the preferred embodiment in appellants' specification, yet 12 of the 29 materials in Table 1 do not come within the scope of the claims allowed by the examiner in the answer. Thus known examples fabricated according to appellants' teaching will not be literally infringed by the Rao, Duncombe and Poole examples.

In Attachment A there are copies of the table of contents and Chapter 3 the 1989 edition of reference 5. Chapter 3 is entitled "Preparative Strategies". In Attachment B there are copies of the table of contents and Chapter 3 of the 1986 edition of reference 5. Chapter 3 in each edition is substantially the same. Since the publication date of the 1986 edition is before appellants filing date, all 29 of the high Tc materials in Table

1 of the Rao article are made according to the general principals of ceramic science as taught by appellants.

Attachment C is a Table of high Tc materials from the "CRC Handbook of Chemistry and Physics" 2000-2001 Edition. Attachment D is a copy of this table with hand written numbers to the left of the materials. There are a total of 42 materials listed in Table 1 (those marked with an asterisk in the table in Attachment D #s 1, 7-13, 16-18, 20, 21, 27, 28, 30, 31 and 41-44) of which 21 do not contain one or more of a rare earth, a group III element or an alkaline earth element. Yet all 42 are made according to the general principals or ceramic science taught by appellants. Two of the 42 materials have a Tc of 25K. Thus a person of skill in the art following appellants' teaching can fabricate materials which do not infringe the claims allowed by the examiner but do not infringe claims not allowed by the examiner.

Table 1 in attachment C list 7 references as the source of the information on the 42 high Tc materials. Those references are listed below. For references 1-5 Attachments E to K, respectively, contain the title page and table of contents of the corresponding book. References 6 and 7 are articles, copies of which are in Attachments J and K respectively.

- 1. Attachment E Ginsburg, D.M., Ed., Physical Properties of High-Temperature Superconductors, Vols. I-III, World Scientific, Singapore, 1989-1992.
- 2. Attachment F Rao, C.N.R., Ed., Chemistry of High-Temperature Superconductors, World Scientific, Singapore, 1991.

- 3. Attachment G Shackelford, J.F., The CRC Materials Science and Engineering Handbook, CRC Press, Boca Raton, 1992, 98-99 and 122-123.
- 4. Attachment H Kaldis, E., Ed., Materials and Crystallographic Aspects of HTc-Superconductivity, Kluwer Academic Publ., Dordrecht, The Netherlands, 1992.
- 5. Attachment I Malik, S.K. and Shah, S.S., Ed., Physical and Material Properties of High Temperature Superconductors, Nova Science Publ., Commack, N.Y., 1994.
- 6. Attachment J Chmaissem, O. et al., Physica C230, 231-238, 1994
- 7. Attachment K Antipov E. V. et al., Physica C215, 1-10, 1993, 231-238, 1994

The is no evidence in these references that the 42 high Tc materials of Attachment C cannot be made following appellants' teaching.

Appellants request the Board to reverse the examiners rejections of claims under 35 USC 112, first paragraph.

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Daniel P. Morris Reg. No. 32,053

(914) 945-3217

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IBM CORPORATION Intellectual Property Law Dept. P.O. Box 218 Yorktown Heights, New York 10598

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BZ & Duncombe

**ATTACHMENT B** 

401001

Technical Notebook

Book IV

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Security Classification:

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•	TARE WEIGHT $A$ , $(X, I)$ $q$ .  SAMPLE WEIGHT $I$ , $(A, C)$ $Q$ .  CELL HOLDER VOLUME, $V_{C}$ $C$	4 001 EAMPLE WEIGHT 16.963 o. ADDED VOLUME, V. 24.85 cc CELL HOLDER VOLUME, V. 25.02 cc
	OPERATIONAL EQUATION $v_p = v_c + \begin{bmatrix} v_A \\ 1 - P_2/P_3 \end{bmatrix}$	OPERATIONAL EQUATION $V_p = V_C + \begin{bmatrix} V_A \\ 1 - P_2/P_3 \end{bmatrix}$
	V <sub>p</sub> = Volume of Powder (cc)	V <sub>p</sub> = Volume of Forder (cc) V <sub>c</sub> = Volume of Sample Cell Holder (cc) V <sub>A</sub> = 'Added Volume
· · · · · · · · · · · · · · · · · · ·	<ul> <li>V<sub>O</sub> = Volume of Sample Cell Holder (cc)</li> <li>V<sub>A</sub> = Added Volume</li> <li>P<sub>2</sub> = Pressure Reading after Pressurizing Cell</li> </ul>	P <sub>2</sub> - Pressure Reading after Pressurizing Coll P <sub>3</sub> - Pressure Reading after Added V <sub>A</sub>
	F <sub>3</sub> = Pressure Reading after Added V <sub>A</sub>	P-3.662 5 R-3.662 8 RUN 3
	R-3.651 R-3.663	8-3,644 <u>RUN 1</u> <u>RÜN 2</u> <u>RUN 1</u>
	*2 18.557 18.568 18.504 *3 5.084 5.085 5.052	8.529 *, <u>5.018</u> <u>5.018</u>
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			•.	Ba	2.04	0.07	3.43
PIL	4	$\mathcal{R}_{\mathfrak{S}}$	Comp	Cu	3.00	0.11	3.67
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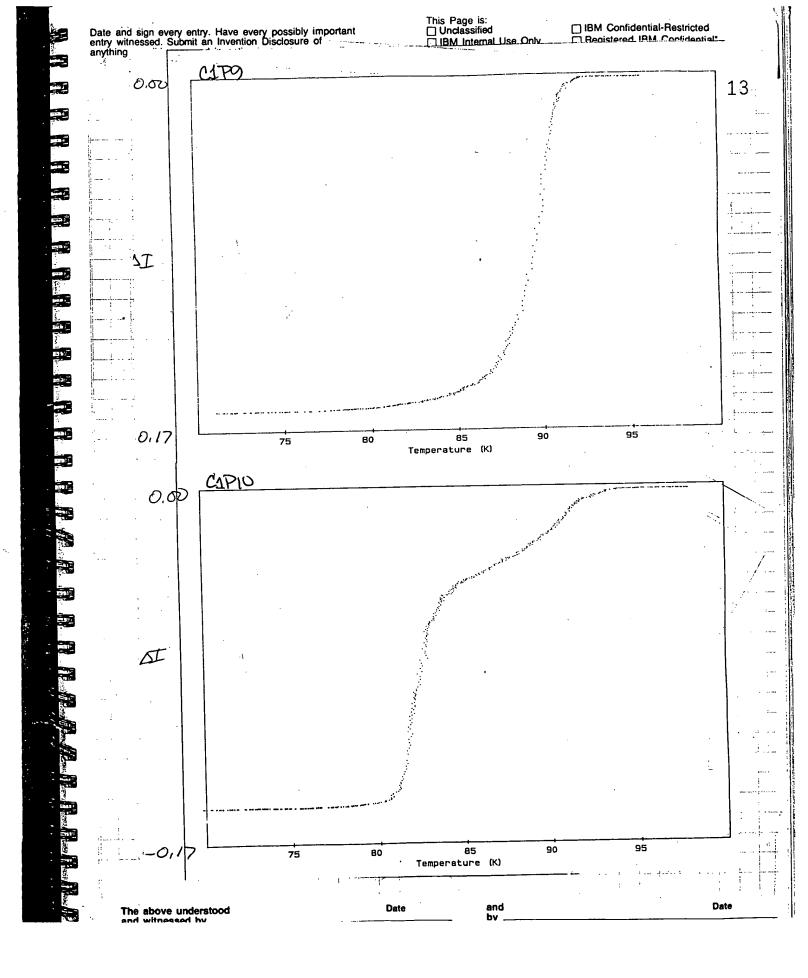
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00 FOR 12/21 → € STIDS RUN 24/HRS → 1000 PSI
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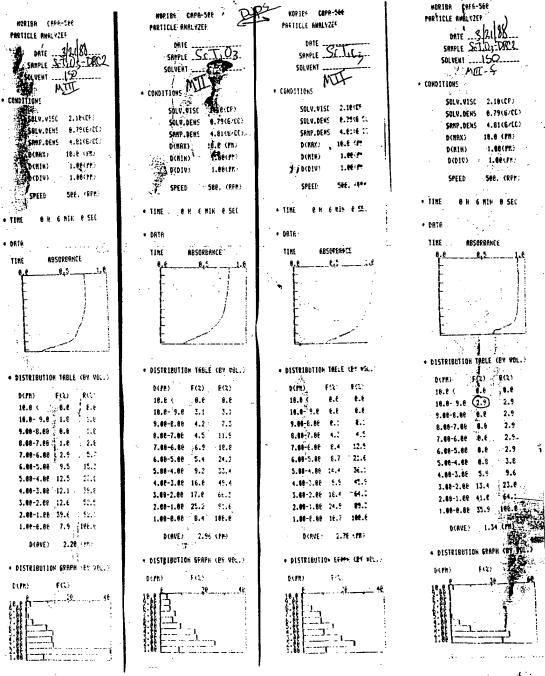
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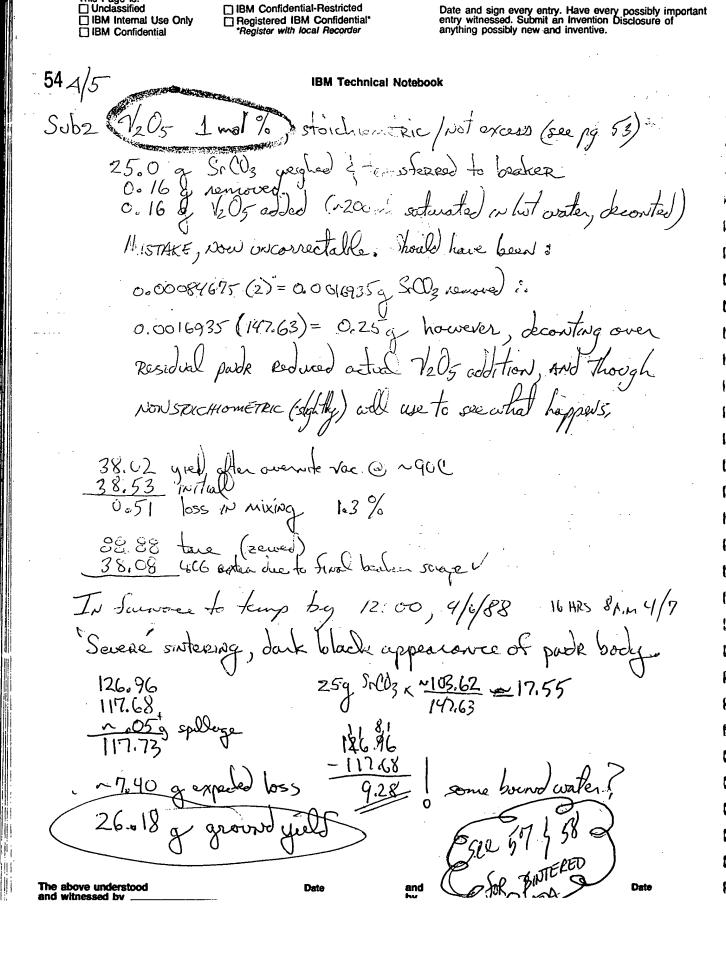
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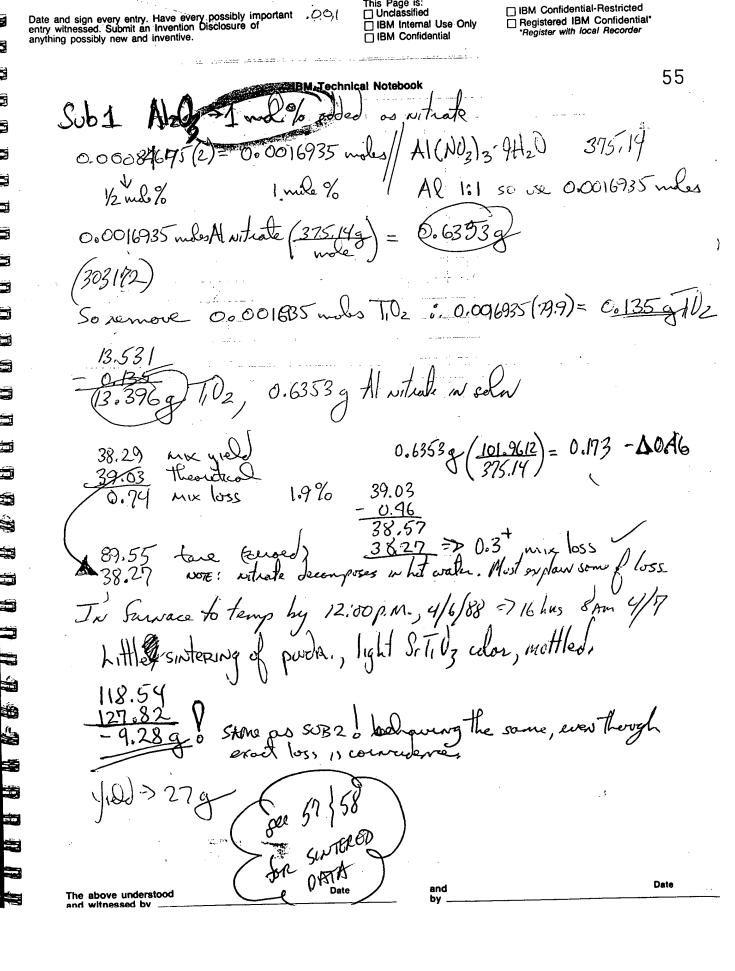


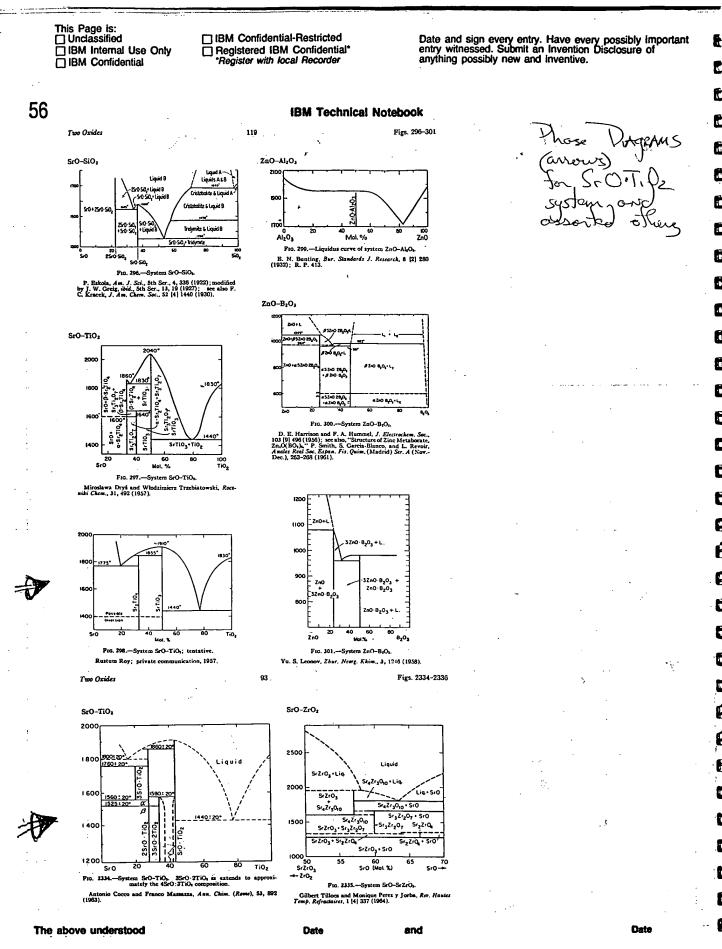
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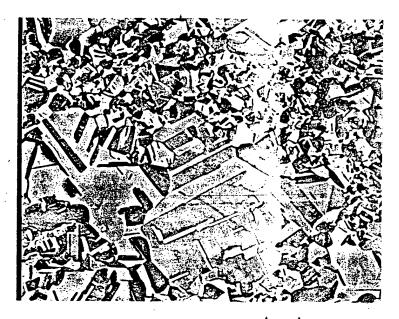
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**Technical Notebook** 

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			Mo ocht hess. 5g	
	6 % 50 10 (3)?	bulance	Conditions: 750C	in N2. unite

n deustry 25 Bi (90) By 61. In al. (assume 9.17g/c. for 2575 fully dense body )
Definitely beautiful Jesised proise or close.
35° Be divide not calculated but fillum objectly smaller is simple deustry to at least some or hydre.
Shell endre of incomplete rotting. Muon third p.
evident do possible evide?

. NOTE: " density

Conditions: 7500 in An/5Hz Compused plat river

sorres: cetting in Nz does not seen to be tetal, possibly order. Films inthumering behaviore. 50/50 mix too Huch lie. Sur parasts apparant. 10% too little lie. No approache cold boss.

NOTES: a cole dons ty 25 B; pund (17%) about loais.
Endre of bella welling, superting only a uning tion
stand providing host downtry due to pullet
bloosting, probably hoppin au in subject to ho
symptom B. Haponychion avident by ught loss

E.	%.B.	& Cu balance	Conditions: Fine 5.11 m Ar	paked pude tro 750c Hz. No con	overite
` <b>v</b> )0	TES! Porce	المعروبة ا	(grey) 3 " " " o 100 seems oup 100 men sph		v
- · · · · · · · · · · · · · · · · · · ·	Ci, they Buted chande	gh 10 jun s	soms to give be some to was better WD	etta overall	Results, Luction

Tolu Constians: 'parked' parke cruelle balonte satura @ 7500 avante w Ag/H2. No composition. ... 25

Results: Cood areas and cetting whol 20 { 25 % samples, however 20% seems botter organil with small on form yours whereas 25% has some voids As well As poses,

20 \ 25% Be simples pressed to 27 500 psc 130statelle. Some collapse of large powerty voids. It the estat of small powerty.

Arreading effect: 0.02 | 0.04 a of Bi-rul ha.

Per out of hicrostructure upon numbering. Porosity drawides.

Very good boking virtually double 20% sample.

Bare: 25% Bi (10g hales) in vitreous boat fast placed in house roccum (dessication) their sintend overwite per stil treatment producing very good looking micro-structure with little porosity.

conQuesoro:	hell me	s. Vocum	A fireston	pren to west.
condusions:	-/Hz. 100 es	xo to compor	adations, his	mever, indicata

Vacuum 25 Bi Run

F.

tocum uno O.K. No evoluci of ropon phase deposition in glass tibe.

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8-11-88

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15

GREEN phase substrate work

have one remaining substrate, n80-90% dense, single phase, sinter 1 13500

1 pressed 0.2", 0.20 pellet et entertie

8-18-88 week sumary

pellet almost totally melts (2 d) with interaction between AlzO3 And 110. B.

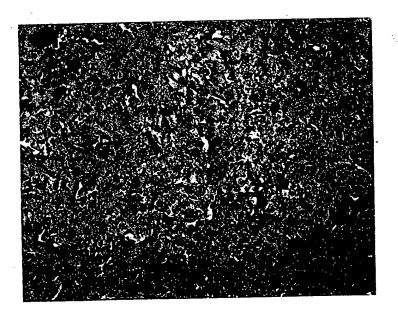
1400 C

pellet Remains \$15 INTECRITY, But large Amount of LIQ forms 20, interaction of lia & And support

lia & still present, though diminished. less interaction. for short sincer time 251 willbd "on"

KOOI

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1292C

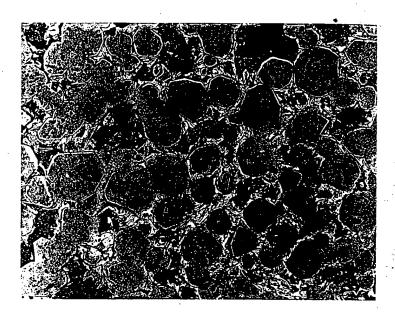
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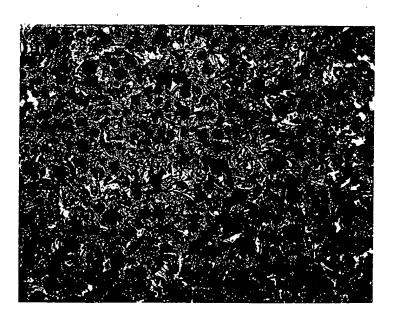
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**IBM Technical Notebook** 

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211 milled 17

1235C

2HPS

1000X



Condusion: sintering @ 1292C on higher creates 2 & material exagginated his & grain growth after prolonged period sintering @ 1235C closs not movice adequate sintering. pollat humains grean as apposed to higher temp, where pellet trens, black (presumably this is not simply source estet, but has chemical basis)

sintering @ 1266C may be optimal.

Purch delinately ow?

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18 Green density - ZIIM 5500 2.56 1.548 0.399	IBM Jethnical Woteb	ook ~7	
	0,7509	3.41-/(6.36)=~~	50%
150 V 1.472 0.389	0.662	3.87/(6.36) ~ (	ø.9
Post >> pellet not good	enut to bother	(6.00) ~ 6 . 20, stuck, etc.	4.5 (1292C) 18H
1265C pellet II (150		2 00 / 1/	,, <u></u> -
pellet cracked on cha	ekwy must re	iso, Temp O.K. thou	4.5
pellet III a furnace	@ 4:10	to temp @ 41:30	) —
SET 1255, TSAMPLE			
2.53 1.286 0.360	0.47 5.	38/(6,00) 🛰 89	9.7 %
8/23/88 15029, pellet N	(second good")		
2.81 1.455 0.488	0.7283	3.86 ~64.3%	
7 2.8 1.283 0.384	0.4964	5.64 494	
15030, pellet I	(edge chipping	dueing isopressing) 0.4	۷,
2.94 1.456 0.457	0.761	3.86 ~64.4%	consistent
4:20 to temp @ 12670	1_		
2.93(5) 1.283 0.4- 2,9 3 good slices	0.517	5.67 u 94.5	265h
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=1	a/a IBM	l Technical Notebook	21
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	2.17	78+8	•
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	Sr 87.62 Sr0	103.6194	
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		·	
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	Lord mindles Pie Uz . 10	2.	2 7688
		17408059 ng	26.1120885
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		.15907880 ng	23.86/82
		Ü	23.86182_ 210.087363 g
	\$ CO3 - 60.00935 \$ CO3 - 147.62935 Ca		210,000 1363 9
	•	CO3 - 100.08935	•
	SKOz: 147.62935 = 1.43 SrO 103.6194	2472693 (26.11208	185) = 37.20259576
	CACO3 : 100.08935 = 1.78	3477926 [9.84193]	5)= 17.56568146
	56.0794	<i>u</i> -	* .
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37. 20259 17. 56568	576 9.30 146 4.3	18.783° 0064894 (0.70188 9 1412037 (0.5602	889)~ 6.528 F
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Cal	6.587	6.59	249.41 (2) <u>256.02</u> 6.61 + -0.01(2)	0.01(2), recovery	
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\ <u></u>	46.5 <b>9</b> (8) 89.12 57.4 <b>9</b> 1/5 57.69	(0.4%)oss	· · · · ·	159,46	
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۱ نلـ	46.59 42.15 post 20 41.44 g loss >	No ENDENSE 7.06 g expected	OF VAPOR  6. 63% convers	ion of no Br loss	· !
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35.13 49.97 49.34	IN furrace (	tube) for 850C,	16h colonation
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9-7 BL 2.15 Sr. 6 C. Ox (ref. data pg 21) 25 2.15 namules B1203 1.00181013 but 2Bi = 1BiO2. 0. BE 0.50090507 mg/mM 1.6 males SrO -> (1.6) (103.6194) = 0.16679104 mg/mM SrO-> SrCO3-> 1.42472693 (0.16579104) = 0.2362 (0696) scale foctor for 50g lot ~60 (60) (0.50090507)= 30.0543 Bi (0.23620696) = 14.1724(0.0795394) = 4.7724 48.9991 Cap. 86 Sro. 14 Co 1 Oz 0.86 (56.0794) = 0.048228284 (1.785.) = 0.0861 0.14 (103.6194) = 0.014506716 (1.42472693)= 0.02066811 1.0 (79.5394) = 0.0795394 scale factor for 50g butch (340) 340 (0.048228284)= 16.398 - 7.027 468 g (less (U2)

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	B1203 melt		la below initial ca	lawation 1
. 1.	CaCO3 deco	mp 825°C)	775.c	
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28	IBM Technical Noteboo	ok 133.66
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133.66 post 7900 89.15 44.51	c 16h brown	wh-black hue, little sintering
132.66 post 825 exterior of	C 16h RICL bl SINTEN MASS TANK	RUN X-RAY IN MORNING.
Calavation I So		CONT Pg 30 >>>
140.92 90.4 <b>9</b> 50.43 /50.52 0.0	7.03 03% loss	2(0.56) = 9.1952 (-0.7.225) 3(0.702) = 4.935 (-0.7.225) -9.32
133.28 90.49 42.79		41.20 : 42.2/42.8 <u>96%</u>
42.43 post gure 132.90	0.8% (055.	Product looked good, little Sintering, black.
Om 110		COLI @ 8000 Then x-ray,

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	/ CAO	excited On	u, Ox Res	TART / method @	1,000 N experimental
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8,29,30/88	LEWIS CO. A.
2	10.3 g collected after granding for possible cal III
5	TO PAGE 32
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	9/14 BLZ10 PLZ1 FLZ	IBM Technical Notebook	31
The Pro-	43 go Buz.15 Sr1.600x	collected after 16h 8250 will awart x-pay tomorro	cal {<100 mest
F 9 9	Desimitely NOT CONVEX	THEO! X-RAY RUN.	still bully ox. X-RO
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第 第 第	38.93 recovery	to crullly	LCPt),
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	<u>کړ</u>	additional peoples, but	precommently
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		Notebook  Lly reported CoSrCu & OOII  valole IN x-ray but multi-component  OOC God safer
126.92 86.66 to 90.26	ere 890C seems	to be temp (+50)
	10/7 Chandra says, temp suggest	880C may be ouset. Final cal.
19/6 925C	16h : X-Ray show	-s
10/7 965C	16 h %26.37 86.66 39.71	oppears as usual, some ninon sintering. All reacting, pattern lossing other peaks, our wereasene intensities of colle
966 C	Swith to Ove out in Removed Ster. )	o 810 when 812 went weld,  sershoot to (006,C for L (minute.  f sample sawit,  s, some Pt exw on I side, hander  ind, preground & replaced tok weekens  ege for the hest,  t= 30 td=10 } continuingation  (temp.)

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THE PR	Sintering Test 830 830	836	pidtemp 1 sample 858 850 egil.	NO SINTERING	appreciation	
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l.17 .0	95 0.278 0.26	2 4.465 69.7	
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	7,30 839 0	as Ts 859 Red	
10/5	4:50 16h 8:30 16h 0.287 0.291	Stable @ 856 Sinter check	
	0.287 0.291	(3.92)	
Pellet warped,	flowed (@approx. da. d KRXV on part of top was 859. Must	us to non-vertical side surface. Paule ter	s) and possibly
PRE 0.94 1.12	the second of th	4.48 69.4	<b>/</b>
pellet boks a	ood @ 852 effer	20 h (overente)	Coep sutering
0.90 1.23	3 ~0.23 0.27	3.3 51%	does not make sense
• • • • • • • • • • • • • • • • • • • •	ys people have see	w such effects	results though
B <sub>2.15</sub> J <sub>1.68</sub> La <sub>7.17</sub> Cu <sub>2</sub> O <sub>84</sub> S "22.12" Avalysis Rosults pg. 33			1000X 120h 850C
"22.12"			850C
Avalysis Results			
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38	IBM Technical Notebook	
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## **IBM Technical Notebook**

P. Husion Pellet Calculations

3.25g/6.36grc = 0.51 cc

0,51 cc x 7.2 g cc = 3.67 g

0.51 cc × 1.86g/cc = 2.48

In preheated RT @ 3:31

951

956

965

4:20

Date

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N RLE	201-8 87 <b>24</b> 201-3 872 201-4 875C	SIMTERT ZAIN 12-3	91.25 9.7 <sup>+</sup> 71.7		-2201 P3 pressid pellet large dre)
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11-12

IBM Technical Notebook

47

201-11 cet what longer flattened and polished.

0011-2201 sawdwhich ~0.353-0363 thich.

> From Survace top to bottom of "weight plate" 1932 @ 462C assuming ~ 6 lbs Son Ram & plate & X-sectional pellet area of 07212/N2 load

Tdiffusion sintering set @ 8600 for ~ 12 hs.

Rel dansity from measurement of 201-11 ~83 %. On suspection of interestal poliched surface numeous buenout-like occlusions present. Some daysee of apen porosity, also.

Pyc. pol. don = 28. % thus 1 attributile to open poresity.

COTT pel done it from measure ~ 89 %. No pyc reading done, 16h sinter @ 975K.

41:30 pr. TO 859C assume start of diffusion sinterinic Plate hieght 13/8" (3/32 exponsion due to TCE from 462CF) No Rt measure mode, but not signify out)

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0011 2201		e de la companya de La companya de la co
~1.43 g ~ 7.53 M	cc ZrOz balls  /3 full	3:00 P.M., 50mls 150 Ruyl.
NOTE: From pottom pg 51	can be seen	this Additive approach will
yield a theoretical	melar comp {	O. 1 M larger in Sr 0.33 M less in CA
	(	0.33M less IN CA
1.e. Strotto enly	(Alcia poop	
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Stoic 1 PRe 2700/27,500		
3.11 1.36 0.486	0.706 4.41	~ 689
0.25 (4)+ 0.75 (7.2)= 6		is, a density cola
RXW. (SINTER) temp to	be 850C	· · · · · · · · · · · · · · · · · · ·
Rellet melter indicenting Kstallyer. Prodomin as in 2201 120h som	lower mp lig vantly 1 lath	of exists in system of later -like of my exigen when growth
		· 
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4:20 P.M. // 4:25 @ temp

OO11-3 placed in pre-heated Rapid temp set @ 951C (Temp=975C)

Sor avernite sintering

No per data on dessity due to irregular shape caused by

pellet crumbling dorning isopressing.

unipress > 6000 ) 150-29,000 PSI white 3.169

9:35 Slow cooling begin . DTsister = 17h @ 8750

Post 2.869 ~0.460 mm thick polis myst have been ~1.3€ estimated density 0.666cc @ 3.19 ~ 4.6€/\$.00 = 93 (may be high) 3.0 4.5 / \$ 90 better

Slice 1 + 0.09" after cleaning / post polish > N/R Slice 2 -> 0.074 0.179

2201-8 1.038 Sia : area = TTD/4 = 0.85 cc² = 0.525 N² 5.75 lbs/.525 N² ~ 11 psi

2201-8 (top)

Pellet consignation @ START ~ 3:55 p.m. thickness - 0.34.cm RAMO > 434 Set point - 800C Duell-12h 1/32@380C

001-3 Ramp > 434 Set point - 8000 Dwell- 12h 1 12/7 Result: No melting, pellets bonded of little deformation.

12/8 After 24h 3250 Anneal no evidence of lia., but bond breaks afterhanding at pellet interface with some "Axw etching" of course pellet surface leaving thin, layer of 2201 (or exa prod) heling.

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~ 0.7019 conversion Sodo	919	97.9982 g	
Estimate ~ 89 g batch R	ecovery ==	97.9982 - <u>8.4468</u> 89.55	Cd <sub>2</sub> /633
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12-8-32		-: 	
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Pellet dagliger than us	ual, 1.75g	max in fiture a	right be considered.
- 1.183 0.715 ce	ACKED, Meas	Att strangers	
12-9 5W-2 900C 8500/39,000		معين	
1.27 1.174 0.382 0	414 3.01	7 61.4	·
3:55 N perhaded furnace > POST 5 MIN 1.24 1.111 0.36 0.	- 4:00 to 7 349 3.	lenp@ 900C	<u>.</u>
15 MIN NO SIGNIFICANT	•	"	
12-12 to temp ~ 10:20 A.M. 12:15 A.M. 14:45.		45 > po slumpi initiated	wy) >> SWIER All 12+30
~2h 1.24 1.055 0.33 (			
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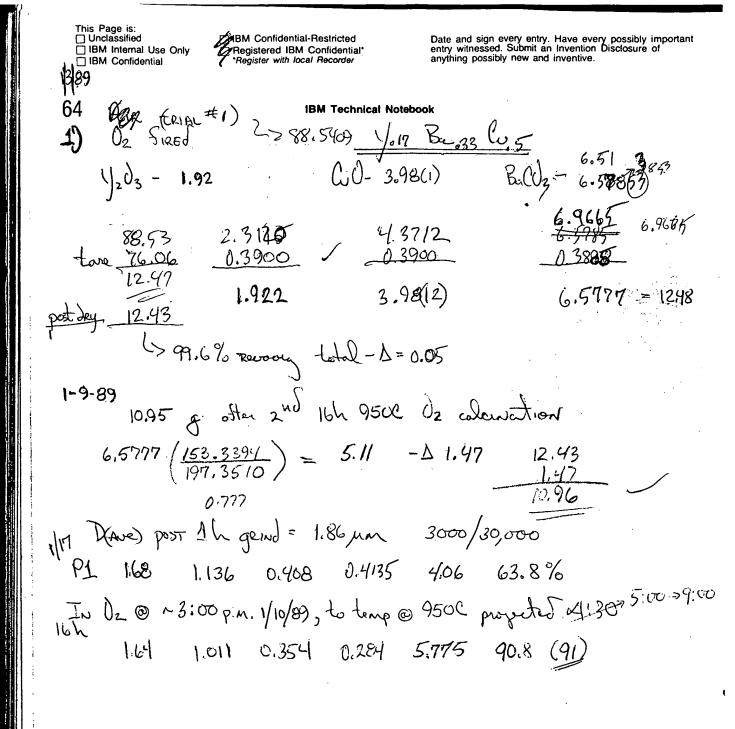
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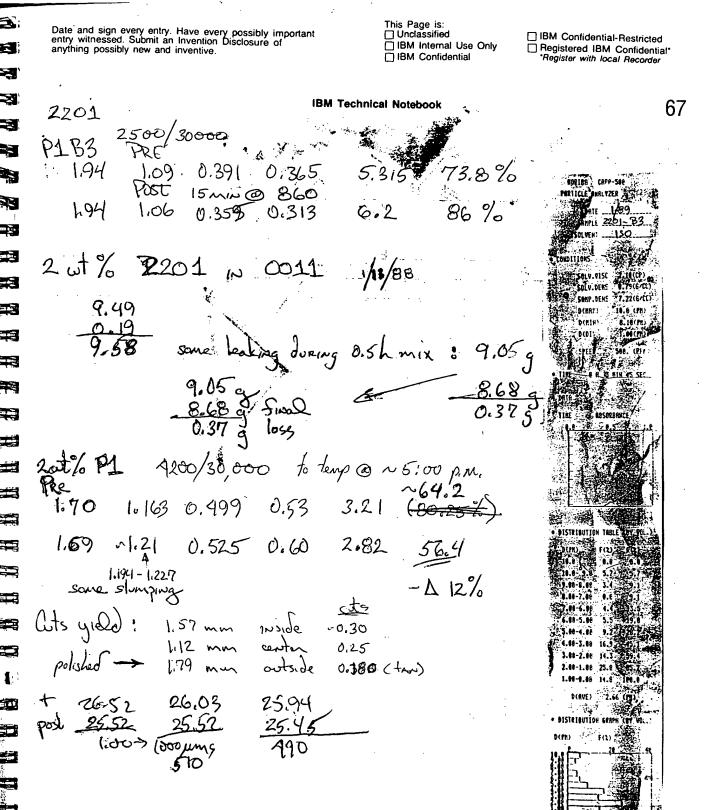
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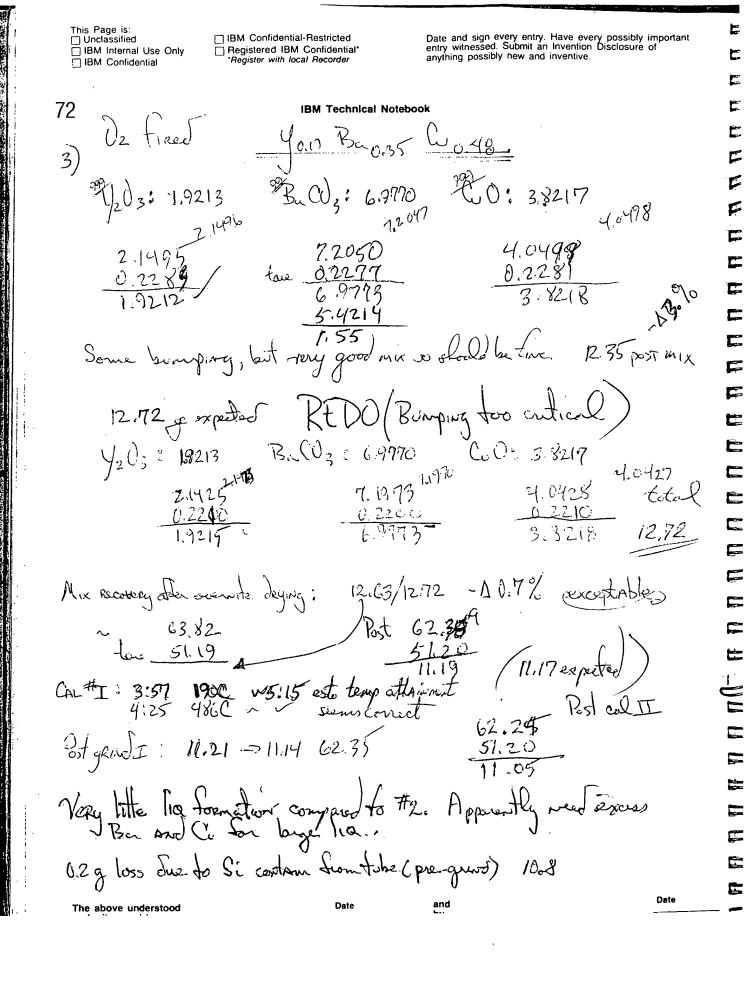
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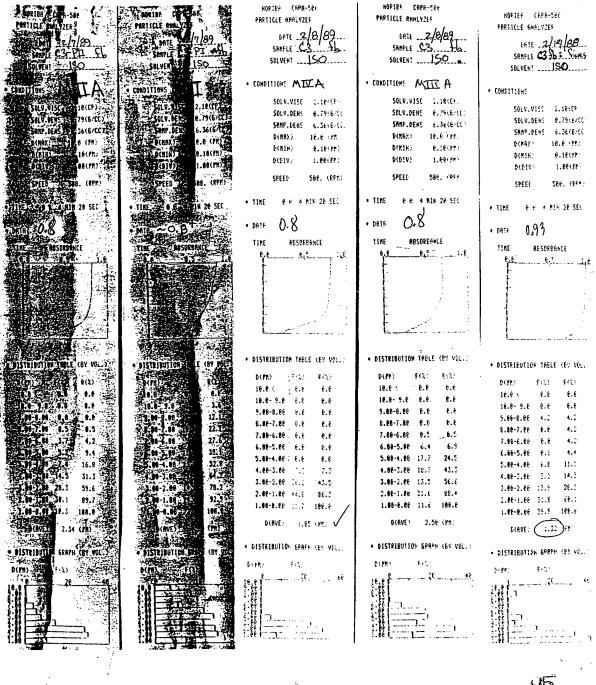
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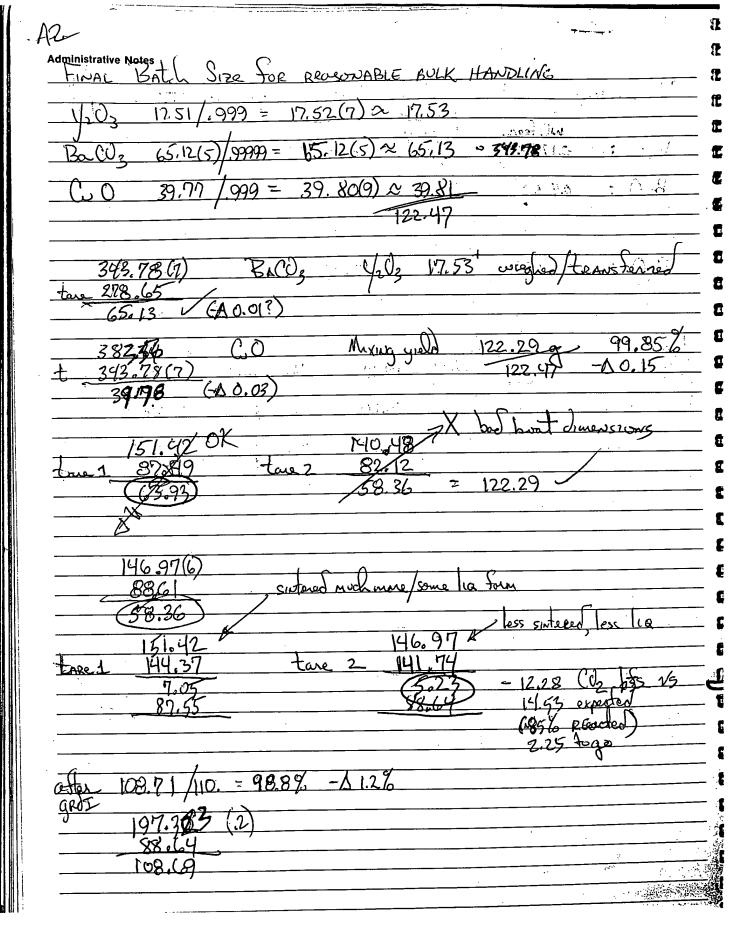
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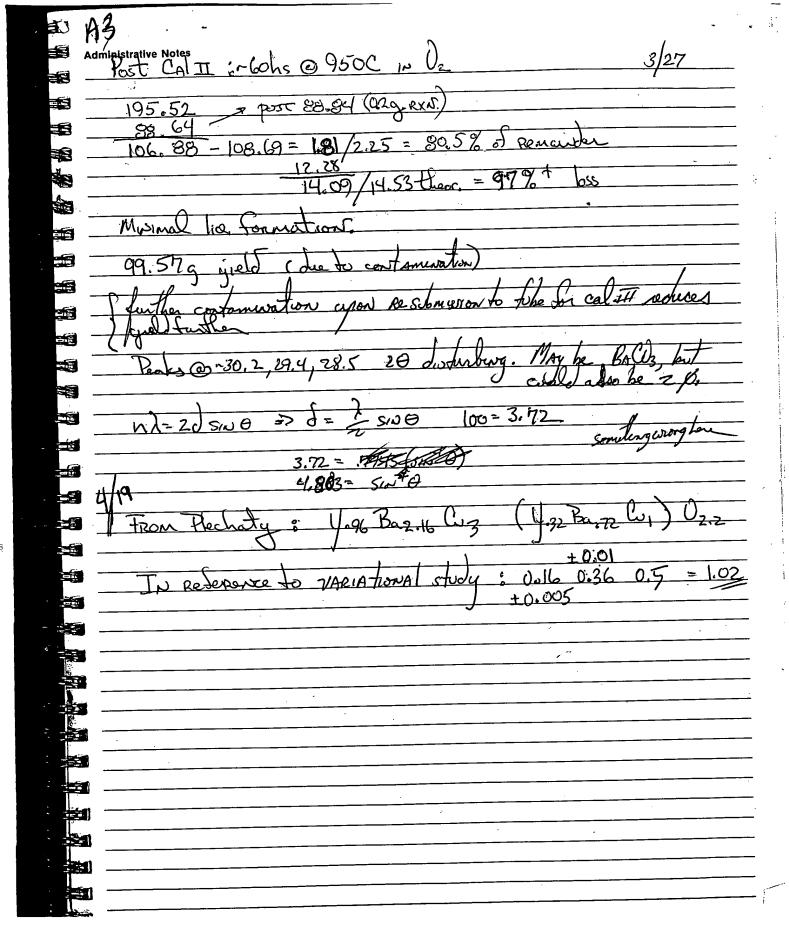
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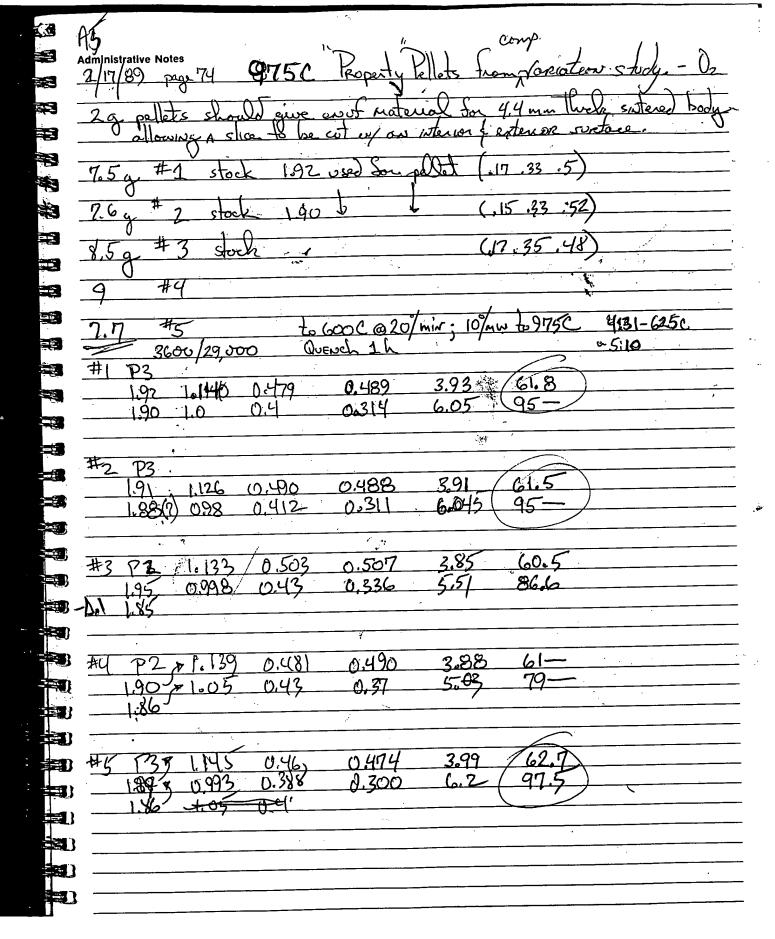
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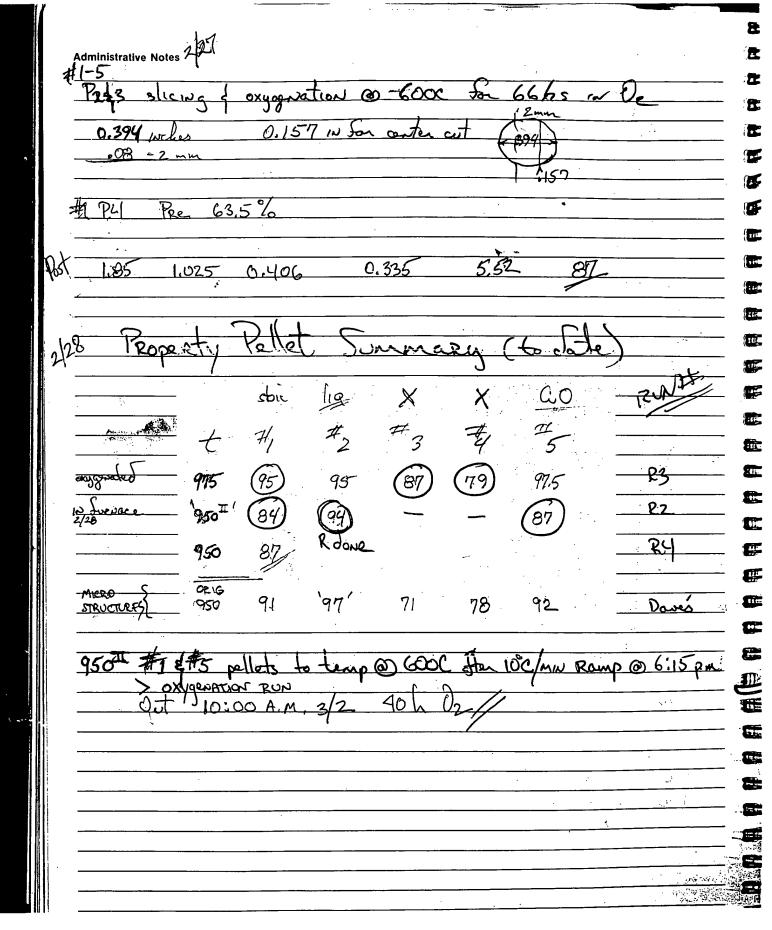
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# BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

Paper	No:	

Application Number: 08/303,561

Filing Date: September 09, 1994

Applicants: Johannes G. Bednorz et al. Examiner M. Kopec

Daniel P. Morris
For Appellant

## FOURTH SUPPLEMENTAL APPELLANTS' REPLY TO THE EXAMINER'S ANSWER

#### **ARGUMENT**

As indicated in Appellant's Request For Postponement of Oral Hearing Scheduled For November 6, 2002, dated November 21, 2002, which was granted by the Order Granting Request To Reset Hearing Date, dated November 6, 2002, attached herewith is a sequential concordance of copies of documentary evidence referred to in Appellants' Brief and Replies which were submitted as attachments and appendixes to papers and affidavits submitted during prosecution. These copies of the concordance is to this Fourth Supplemental Appellants' Reply To The Examiner's Answer. The first pages of the Concordance is marked Concordance and Contains a Table of Contents which lists the documents in the order of first appearance in the Brief and Replies. All page are identified where the document occurs in the Brief and Reply Briefs. Documents in Appendix of The Brief and Reply Briefs are not included in the Concordance.

#### ADDITIONAL REMARKS CITING PORTIONS OF THE FILE HISTORY

Claims 24-26, 88-90, 96-102, 109-113, 129-131, 134, 135, 139-142, 145, 149-151, 158, 159, 164-166, 169-170 and 174-177 of the present application have been rejected as not enabled under 35 U.S.C. 112, first paragraph. Appellants disagree for the reasons previously noted. Appellants in addition point out the following.

The present application is a Continuation of 08/060,470 filed on 05/11/93, which is a Continuation of 07/875,003 filed on 04/24/92, which is a Divisional of 07/053,307 filed on 05/22/87 all now abandoned.

In the 07/053,307 ancestral application composition of matter claims where presented for examination. A copy of the Final Rejection referred to below in this application is in Attachment A of this paper.

In the 07/053,307 ancestral application composition of matter, claims 1 through 11 inclusive, 27 through 35 inclusive, 40 through 54 inclusive, 60 through 63 inclusive, and 65 through 68 were finally rejected under 35 U.S.C. 102(b) or in the alternative under 35 U.S.C. 103 as unpatentable over each of a publication by Shaplygin et al. in the Russian Journal of Inorganic chemistry, volume 24, pages 820-824 (1979) ("the Shaplygin et al. publication"); a publication by Nguyen et al. in the Journal of Solid State Chemistry, volume 39, pages 120-127 (1981) ("the Nguyen et al. publication"); a publication by Michel et al. in the Materials Research Bulletin, volume 20, pages 667-671 (1985) ("the 1985 Michel et al. publication"); and a publication by Michel and Raveau in the Revue de Chimie Minerale, volume 21, pages 407-425 (1984) ("the 1984 Michel and Raveau publication"). See the final rejection dated 4-25-1991 in the 07/053,307 ancestral application.

In the 07/053,307 ancestral application, claims 1, 2, 5 through 11 inclusive, 40 through 44 inclusive, 46, 48, 51 through 54 inclusive, 60, 62, and 66 were finally rejected under 35 U.S.C. 102(b) or in the alternative under 35 U.S.C. 103 as unpatentable over a

publication by Perron-Simon et al. in <u>C. R. Acad. Sc. Paris</u>, volume 283, pages 33 through 35 (12 July 1976) ("the Perron-Simon et al. publication"); a publication by Mossner and Kemmler-Scak in the <u>Journal of the Less-Common Metals</u>, volume 105, pages 165 through 168 (1985) ("the Mossner and Kemmler-Sack publication"), a publication by Chincholkar and Vyawahare in <u>Thermal Analysis</u> 6th, volume 2, pages 251 through 256 (1980) ("the Chincholkar and Vyawahare publication"); a publication by Ahmad and Sanyal in <u>Spectroscopy Letters</u>, volume 9, pages 39 through 55 (1976) ("the Ahmad and Sanyal publication"); a publication by Blasse and Corsmit in the <u>Journal of Solid State Chemistry</u>, volume 6, pages 513 through 518 (1973) ("the Blasse and Corsmit publication"); United States Patent No. 3,472,779 to Kurihara et al. ("the Kurihara et al. '779 patent"); a publication by Anderton and Sale in <u>Powder Metallurgy</u> No. 1, pages 14 through 21 (1979) ("the Anderton and Sale publication"). (See the final rejection dated 4-25-1991).

In the 07/053,307 ancestral application the Examiner asserted that the cited references appeared to disclose materials, which inherently provided superconductive properties and assertedly therefore, rendered the claims unpatentable. Appellants rebutted the Examiner's reasons for rejection based on limitations in the claims directed to Appellants' new discovery of the superconductive properties of these materials.

The claims of the present application are directed to methods of flowing a superconducting current in a superconductive composition of matter having a transition temperature greater than 26 K. This is Applicants' discovery for which they received the 1987 Nobel Prize in Physics. The Examiner in the 07/053,307 ancestral application stated by the 35 U.S.C. 102 and 103 rejections therein that persons of skill in the art knew how to make the compositions of matter based on the references cited therein. In that same final rejection the Examiner states at page 4 thereof "these materials appear to be identical to those presently claimed except that the superconductive properties are not disclosed." Applicants discovered the superconductive properties and in the present application are claiming methods using this property. Thus, by the Examiner's reasoning all of the present claims are fully enabled because the Examiner has stated

that the compositions of matter recited in the claims can be made with the knowledge of a person of skill in the art prior to Applicant's filing date. Thus the Examiner, in the 07/053,307 ancestral application, agrees with the Appellants' Arguments and the Affidavits of Shaw, Duncombe, Tsuei, Dinger and Mitzi submitted by Appellant in support of their position that all their claims are enabled. In view thereof, Applicants request the Board to reverse the rejection of the claims under 35 U.S.C. 112, first paragraph as not enabled.

Section 104(b)(3) of 37 CFR states "[i]n rejecting claims the examiner may rely upon admissions by the applicant ... as to any matter affecting patentability". Thus, if Appellant in rebutting the 35 USC 112 rejections made statements adverse to their interests in regards to rebutting the rejections under 35 USC 102 and 103, the Examiner could use these statements to assert that Appellants admitted that their invention was anticipated or obvious. Likewise, if Appellant in rebutting the 35 USC 102 and 103 rejections made statements adverse to their interest in regards to rebutting the rejections under 35 USC 112, the Examiner could use these statements to assert that Appellants admitted that their claims were not enabled or were indefinite. Section 104(b)(3) does not explicitly apply to the Examiner. Not to apply the rational of 37 CFR 104(b)(3) to the Examiner to find that the necessary consequences of the Examiners 35 USC 102 and 103 rejections of the composition of matter claims of the 07/053,307 are that all of the Appellants' claims are enabled would be manifestly unfair and inequitable.

Claims 86, 87, 96-108, 112, 113, 117, 118, 122, 123, 127, 128 and 147 have been rejected as indefinite under 35 U.S.C. 112, second paragraph.

These claims have been rejected under 35 USC 112, second paragraph, as indefinite for using language of the type "rare earth like" and "pervskite-like", etc. As previously stated the Examiner has arbitrarily rejected Appellants' claims without providing a reason for why Appellants' terms are indefinite while similar terms are not indefinite in the claims of many issued patents. Applicants note that article incorporated by reference at page 6 of the specification were published in September 1986 (which lead

to Appellants' Nobel Prize) and the present application was filed in May 1987 thereby clearly making this terminology part the high Tc superconductor art. As shown this is the vernacular of the field and well understood by persons of skill in the art. Appellants request the Board to reverse the rejections of claims 86, 87, 96-108, 112, 113, 117, 118, 123, 127 and 147 under 35 U.S.C. 112, second paragraph.

Applicants' invention is a pioneering invention. "The Supreme Court in Westinghouse v. Boyden Power Brake Co., 170 U.S. 537, 562 (1898), characterized a pioneering invention as "a distinct step in the progress of the art, distinguished from a mere improvement or perfection of what had gone before." Texas Instruments ICC 6 USPQ 2d 1886 (CAFC 1988). Applicants received the 1987 Nobel Prize in Physics for there discovery of superconductivity at Tc greater that or equal to 26'K which is about 8'K higher than the highest Tc previously known. Even though others following Applicants' teaching identified compositions having Tc more than 100'K greater than 26'K only Applicants have received a Nobel Prize for this subject matter. This is because the others followed applicants' teaching to identify these other compositions.

Appellants request the Board to reverse the examiners rejections of claims under 35 USC 112, first paragraph and second paragraph.

Please charge any fee necessary to enter this paper to deposit account 09-0468.

Respectfully subplitted

Dr. Daniel P. Morris, Esq.

Reg. No. 32,053 (914) 945-3217

IBM CORPORATION
Intellectual Property Law Dept.
P.O. Box 218
Yorktown Heights, New York 10598

### **ATTACHMENT A**



SERIAL NUMBER

14. Other

FILING DATE

FIRST NAMED INVENTOR

### UNITED STAT DEPARTMENT OF COMMERCE Patent and Trademark Office

Address : COMMISSIONER OF PATENTS AND TRADEMARKS Washington, D.C. 20231

ATTORNEY DOCKET NO.

07/053,307 05/22/87 BEDNORZ	J Y0987-074
	EXAMINER
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J. DAVID ELLETT IBM INTELLECTUAL PROPERTY LAW DEPT. P.O. BOX 218	ART UNIT PAPER NUMBER
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This application has been examined Responsive to communication filed on_	This action is made final.
	nth(s), days from the date of this letter.
Failure to respond within the period for response will cause the application to become at	25 October 193
Part I THE FOLLOWING ATTACHMENT(S) ARE PART OF THIS ACTION:	
1. Notice of References Cited by Examiner, PTO-892.	Notice re Patent Drawing, PTO-948.
<ol> <li>Notice of Art Cited by Applicant, PTO-1449.</li> <li>Information on How to Effect Drawing Changes, PTO-1474.</li> </ol>	Notice of Informal Patent Application, Form PTO-152
Pert II SUMMARY OF ACTION	
1. Claims	are pending in the application.
Of the above, claims 12-26 36-39 55-595-	Are withdeawn from consideration.
2. Claims	have been cancelled.
	MA .
3. L Claims	are allowed. Z
4. K Claims 1-11, 17-35, 40-54, 60-63 + 65	6 0 are relected.
5. Claims	are objected to.
6. Claims	are subject to restriction or election requirement.
7. This application has been filed with informal drawings under 37 C.F.R. 1.85	which are acceptable for examination purposes.
Formal drawings are required in response to this Office action.	- Transition of the Application を使いません。 - Transition of the Application を使いません。
9. The corrected or substitute drawings have been received on	Under 37 C.F.R. 1.84 these drawings
1. In corrected or substitute drawings have been received on	
10. The proposed additional or substitute sheet(s) of drawings, filed on	has (have) been approved by the
11. The proposed drawing correction, filed, has been	approved; Casapproved (see explanation).
12. Acknowledgement is made of the claim for priority under U.S.C. 119. The o	•
been filed in parent application, serial no; filed or	
13. Since this application apppears to be in condition for allowance except for fo accordance with the practice under Ex parte Quayle, 1935 C.D. 11; 453 O.G.	rmal matters, prosecution as to the ments is closed in . 213.

1. Applicant's election with traverse of Group I in Paper No. 22 is acknowledged. The traversal is on the ground(s) that the claims of Groups I, II and III are not distinct. This is not found persuasive because the Examiner maintains that the superconductive product, process of making and method of use are directed to patentally distinct inventions. Although there are broad "process" and "method" claims that appear to encompass a great deal of subject matter, the limitations in the dependent claims distinguish the claims of the Groups I, II and III.

The requirement is still deemed proper and is therefore made FINAL.

- 2. The objection to the specification and objection of claims 1-11, 27-35, 40-54, 60-63 and 65-68 under 35 USC 112, first paragraph, is maintained.
- 3. The following is a quotation of the first paragraph of 35 U.S.C. § 112:

The specification shall contain a written description of the invention, and of the manner and process of making and using it, in such full, clear, concise, and exact terms as to enable any person skilled in the art to which it pertains, or with which it is most nearly connected, to make and use the same and shall set forth the best mode contemplated by the inventor of carrying out his invention.

The specification is objected to under 35 U.S.C. § 112, first paragraph, as failing to provide an enabling disclosure commensurate with the scope of the claims.

4. The Applicants assert that "the scope of the claims as presently worded is reasonable and fully merited" (page 17 of

Serial No. 07/53,307 Art Unit 115

response). The Examiner disagrees. The present claims are broad enough to include a substantial number of inoperable compositions.

- 5. The rejection of claims 1-11, 27-35, 40-54, 60-63 and 65-68 under 35 USC 112, second paragraph is maintained.
- 6. Claims 1-11, 27-35, 40-54, 60-63 and 65-68 are rejected under 35 U.S.C. § 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention.
- 7. The amended term "rare earth-like" is vague. With respect to the lack of stoichiometry, Applicants argue the superconductive properties can be measured as the composition is varied. This is unpersuasive because the present claims broad enough to require an undue amount of experimentation.
- 8. The Examiner maintains that the term "doping" is vague. Neither the claim or the specification discuss the limits of the effective amounts of doping.
- 9. The Applicants assert that a discussion of "electron-phonon interactions to produce superconductivity" is found in the specification. The Examiner maintains that the term is not adequately explained. The specification fails to teach how one determines how to enhance the "electron-phonon" interactions?
- 10. The term "at least four elements" is indefinite considering the number of elements in the periodic table.

Serial No. 07/53,307 Art Unit 115

- 11. The rejection of claims 1-11, 27-35, 40-54, 60-63 and 65-68 under 35 USC 102/103 is maintained.
- 12. Claims 1-11, 27-35, 40-54, 60-63 and 65-68 are rejected under 35 U.S.C. § 102(b) as anticipated by or, in the alternative, under 35 U.S.C. § 103 as obvious over each of Shaplygin et.al., Nguyen et.al., Michel et.al. (Mat. Res. Bull. and Revue de Chimie).
- 13. The Applicants argue that "no prima facie case has been made that the composition anticipates or renders obvious the subject matter" (page 28 of response). The Examiner maintains that these materials appear to be identical to those presently claimed except that the superconductive properties are not disclosed. Applicants have not provided any evidence that the compositions of the cited references are in any way excluded by the languange of the present claims, i.e. Applicants have failed to show that these materials are not superconductive. Applicant's composition claims do not appear to exclude these materials.
- 14. Applicants further argue that under United States patent law they are entitled to claim compositions which might happen to overlap a portion of the concention ranges broadly recited in the cited references. "The broad statement of a concentration range in the prior art does not necessarily preclude later invention within the concentration range" (page 29 of response). The Examiner fails to understand how Applicant's incredibly broad claims, some of

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Serial No. 07/53,307 Art Unit 115

which require only the presence of a "doped transition metal oxide" in anyway fall "within" the scope of (see claim 42), The cited references compositions disclosed in the prior art. disclose very specific compostions that not only fall within the scope of the claims, but appear to be identical to being specification the disclosed in compositions superconducting. The Examiner maintains that these materials are claim render the inherently superconductive and therefore unpatentable.

- regarding the "question of non-analogous art" and the assertion the cited prior art is irrevelant to the present claim, the Examiner maintains that for the present "composition" claims the references directed to what appear to be identical materials (both in composition and inherent properities) are clearly relevant. The cited <u>individual</u> disclosures appear to be sufficient to maintain the rejection, the Examiner is not relying on any secondary references to modify the teachings in the references.
  - 16. The rejection of claims 1-2, 5-11, 40-44, 46, 48, 51-54, 60, 62 and 66 under 35 USC 102/103 is maintained.
- 17. Claims 1-2, 5-11, 40-44, 46, 48, 51-54, 60, 62 and 66 are rejected under 35 U.S.C. § 102(b) as anticipated by or, in the alternative, under 35 U.S.C. § 103 as obvious over each of Perron-

Serial No. 07/53,307

Art Unit 115

Simon et.al., Mossner et.al., Chincholkar et.al., Amad et.al., Blasse et.al., Kurihara et.al. and Anderton et.al.

- 18. This rejection is maintained for the reasons set forth in the previous paragraphs. The Examiner maintains that the cited references appear to disclose materials which inherently provide superconductive properties and therefore renders the present claims unpatentable.
- 19. THIS ACTION IS MADE FINAL. Applicant is reminded of the extension of time policy as set forth in 37 C.F.R. § 1.136(a).

A SHORTENED STATUTORY PERIOD FOR RESPONSE TO THIS FINAL ACTION IS SET TO EXPIRE THREE MONTHS FROM THE DATE OF THIS ACTION. IN THE EVENT A FIRST RESPONSE IS FILED WITHIN TWO MONTHS OF THE MAILING DATE OF THIS FINAL ACTION AND THE ADVISORY ACTION IS NOT MAILED UNTIL AFTER THE END OF THE THREE-MONTH SHORTENED STATUTORY PERIOD, THEN THE SHORTENED STATUTORY PERIOD WILL EXPIRE ON THE DATE THE ADVISORY ACTION IS MAILED, AND ANY EXTENSION FEE PURSUANT TO 37 C.F.R. § 1.136(a) WILL BE CALCULATED FROM THE MAILING DATE OF THE ADVISORY ACTION. IN NO EVENT WILL THE STATUTORY PERIOD FOR RESPONSE EXPIRE LATER THAN SIX MONTHS FROM THE DATE OF THIS FINAL ACTION.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to John Boyd whose telephone number is (703) 308-3314.

Any inquiry of a general nature or relating to the status of this application should be directed to the Group receptionist whose telephone number is (703)-308-0661.

Serial No. 07/53,307

Art Unit 115

J.Boyd

April 24, 1991

PAUL LIEBERMAN
SUPERVISORY PRIMARY EXAMINER
ART UNIT 115

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- 1 European Patent Application 0 275 343 A1. Pages 7, 11, 12, 13, 14, 16 and 18.
- 2 Applicants' paper submitted April 11, 1996. Pages 7 and 26.
- Z. Phys. B. Condensed Matter 64, pages 189-193, (1986).
  Pages 10, 11, 16, 17, 23, 24, 30, 31, 33, 36, 37, 49, 50, 74, 75, 94 and 109.
  Reply Page 64.
- 4 Applicants' Response filed March 6, 1997 (Paper #59). Page 17.
- Attachment C of Applicants' Response dated September 25, 1995. (Book by M. von Laue).

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- 6 Asahi Shinbaum Article.
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- 7 Attachment K of Applicants' Response dated December 27, 1998. Pages 23 and 24.
- 8 Applicants' Response dated December 18, 1998. Page 24.
- 9 Applicants' Argument filed January 3, 1996 (Paper #50). Page 26.
- 10 Affidavit of Chang C. Tsuei dated January 2, 1996 and filed January 3, 1996 (Paper #52).

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- 10A Declaration of co-inventors, J. G. Bednorz and K. A. Mueller dated March 21, 1988, mailed to USPTO on June 22, 1988. Page 41.
- 11 Declaration of Alexis P. Malozemoff signed March 30, 1988. Page 41.
- Declaration of Cheng-Chung John Chi dated March 29, 1988 and signed March 30, 1988.

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- Declaration of Sung IL Park dated March 29, 1988 and signed March 30, 1988. Page 42.
- Declaration of Chang C. Tsuei dated March 29, 1988 and signed March 30, 1988.

  Page 43.
- Declaration of Dr. Richard L. Green dated March 29, 1988 and signed March 30, 1988.
  Pages 43 and 44.
- 15A Article of B. Raveau, Mat. Res. Bull., Vol. 20, pages 667-671, 1985. Page 58.
- 15B Article of C. Michel et al., Revue De Chimie Minerale, Vol. 21, No. 4, pages 407-425, (1984).
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- Affidavit of David B. Mitzi dated and signed on December 15, 1998. Pages 60, 61, 64, 66, 68, 71, 74, 93, 94, 96, 98 and 100. Reply Pages 25, 38, 51 and 58.
- Affidavit of Timothy Dinger dated December 15, 1998 and signed December 16, 1998.
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- 19 Affidavit of Thomas M. Shaw dated December 15, 1998 and signed December 14, 1998.
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- 20 Affidavit of Peter R. Duncombe dated and signed on December 18, 1998. Pages 60, 61, 64, 66, 68, 71, 74, 84, 85, 89, 93, 94, 95, 96, 98 and 100. Reply Pages 25, 38, 51, 58, 59 and 66.
- 21 Book of Charles P. Poole, Jr.
  Pages 60, 61, 64, 66, 71, 74, 95, 96, 98, 99, 100 and 107.
  Reply Pages 3, 21, 25, 38, 39, 46, 51, 54, 55, 56, 57, 58, 59, 65 and 66.

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22 Affidavit of James W. Leonard Under 37 CRF 1.132 submitted December 15, 1998. Page 75. Reply Page 61. 23 Attachment A of Applicants' Response dated May 14, 1998. Pages 96 and 97. 24 Attachment H of Applicants' Response dated November 28, 1997. Pages 96 and 97. 25 Attachment A of Applicants' Response dated November 28, 1997. Page 105. 26 Attachment B of Applicants' Response dated November 28, 1997. Page 105. 27 Attachment C of Applicants' Response dated November 28, 1997. Page 106. 28 Attachment D of Applicants' Response dated November 28, 1997. Page 106. Attachment E of Applicants' Response dated November 28, 1997. 29 Page 106. Attachment F of Applicants' Response dated November 28, 1997. 30 Page 106. 31 Attachment I of Applicants' Response dated November 28, 1997. Page 107. 32 Attachment A of Applicants' Response dated December 27, 1997. Page 107. 33 Attachments B through J of Applicants' Response dated December 27, 1997. Pages 107 and 108. 34 Attachment M of Applicants' Response dated November 28, 1997. Page 109. 35 Attachment A of Applicants' Response dated December 22, 1998.

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#### **EUROPEAN PATENT APPLICATION**

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- Representative: Rudack, Günter O., Dipi.-ing. IBM Corporation Säumerstrasse 4 CH-8803 Rüschlikon(CH)
- New superconductive compounds of the K2NIF4 structural type having a high transition temperature, and method for fabricating same.
- The superconductive compounds are oxides of the general formula RE2.,AE,TM.O4., wherein RE is a rare earth, AE is a member of the group of alkaline earths or a combination of at least two member of that group, and TM is a transition metal, and wherein x < 0.3 and 0.1  $\le y \le 0.5$ . The method for making these compounds involves the steps of coprecipitating aqueous solutions of the respective nitrates of the constituents and adding the coprecipitate to oxalic acid, decomposing the precipitate and causing a Solid-state reaction at a temperature between 500 and 1200°C for between one and eight hours, formming pellets of the powdered product at high presgure, sintering the pellets at a temperature between 500 and 1000°C for between one half and three nours, and subjecting the pellets to an additional annealing treatment at a temperature between 500 and 1200°C for between one half and five hours in a protected atmosphere permitting the adjustment of the oxygen content of the final product.

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0 275 343

## NEW SUPERCONDUCTIVE COMPOUNDS OF THE K,NIF, STRUCTL'RAL TYPE HAVING A HIGH TRANSITION TEMPERATURE, AND METHOD FOR FADRICATING SAME

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#### Field of the Invention

The invention relates to a new class of superconductors, in particular to components of the K<sub>2</sub>NiF<sub>2</sub> type of structure having superconductor properties below a relatively high transition temperature, and to a method for manufacturing those compounds.

#### Background of the Invention

Superconductivity is usually defined as the complete loss of electrical resistance of a material at a well-defined temperature, it is known to occur in many materials: About a quarter of the elements and over 1000 alloys and components have been found to be superconductors. Superconductivity is considered a property of the metallic state of the material, in that all known superconductors are metallic under the conditions that cause them to superconduct. A few normally non-metallic materials, for example, become superconductive under very high pressure, the pressure converting them to metals before they become superconductors.

Superconductors are very attractive for the generation and energy-saving transport of electrical power over long distances, as materials for forming the coils of strong magnets for use in plasma and nuclear physics, in nuclear resonance medical diagnosis, and in connection with the magnetic levitation of fast trains. Power generation by thermonuclear fusion, for example, will require very large magnetic fields which can only be provided by superconducting magnets. Certainly, superconductors will also find application in computers and high-speed signal processing and data communication.

While the advantages of superconductors are quite obvious, the common disadvantage of all superconductive materials so far known lies in their very low transition temperature (usually called the critical temperature  $T_c$ ) which is typically on the order of a few degrees Kelvin. The element with the highest  $T_c$  is niobium (9.2 K), and the highest known  $T_c$  is about 23 K for NB<sub>3</sub>Ge at ambient pressure.

Accordingly, most known superconductors require liquid helium for cooling and this, in turn, requires an elaborate technology and as a matter of principle involves a considerable investment in cost and energy.

It is, therefore, an object of the present inven-

tion to propose compositions for high- $T_{\rm e}$  superconductors and a manufacturing method for producing compounds which exhibit such a high critical temperature that cooling with liquid helium is obviated so as to considerably reduce the cost involved and to save energy.

The present invention proposes to use compounds having a layer-type structure of the kind known from potassium nickel fluoride K<sub>2</sub>NiF<sub>4</sub>. This structure is in particular present in oxides of the general composition RE<sub>2</sub>TM.O<sub>4</sub>, wherein RE stands for the rare earths (lanthanides) and TM stands for the so-called transition metals, it is a characteristic of the present invention that in the compounds in question the RE portion is partially substituted by one member of the alkaline earth group of metals, or by a combination of the members of this alkaline earth group, and that the oxygen content is at a dollicit.

For example, one such compound that meets the description given above is lanihanum copper oxide La<sub>2</sub>CuO<sub>4</sub> in which the lanthanum -which belongs to the IIIB group of elements-is in part substituted by one member of the neighboring IIA group of elements, viz. by one of the alkaline earth metals (or by a combination of the members of the IIA group), e.g., by barium. Also, the oxygen content of the compound is incomplete such that the compound will have the general composition La<sub>2</sub>,  $_4$ Ba $_8$ CuO<sub>4,y</sub>, wherein x ≤ 0.3 and y < 0.5.

Another example for a compound meeting the general formula given above is lanthanum nickel oxide wherein the tanthanum is partially substituted by strontium, yielding the general formula La<sub>2</sub>. <sub>4</sub>Sr<sub>n</sub>NiO<sub>4-y</sub>. Still another example is cerium nickel oxide wherein the cerium is partially substituted by calcium, resulting in Ce<sub>2-x</sub>Ca<sub>x</sub>NiO<sub>4-y</sub>.

The following description will mainly refer to barium as a partial replacement for the lanthanum in a La<sub>2</sub>CuO<sub>4</sub> compound because it is the Ba-La-Cu-O system which is, at least at present, the best understood system of all possible. Some compounds of the Ba-La-Cu-O system have been described by C. Michel and B. Raveau in Rev. Chim. Min. 21 (1984) 407, and by C. Michel, L. Er-Rakho and B. Raveau in Mat. Res. Bull., Vol. 20, (1985) 687-871. They did, however, not find nor try to find, superconductivity.

Experiments conducted in connection with the present invention have revealed that high-T<sub>c</sub> superconductivity is present in compounds where the rare earth is partially replaced by any one or more of the other members of the same IIA group of elements, i.e. the other alkaline earth metals. Ac-

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tually, the  $T_e$  of La2CuO  $_{\rm e,v}$  with  $Sr^2$  is higher and is superconductivity-induced diamagnetism larger than that found with  $Ba^2$  and  $Ca^2$ 

As a matter of fact, only a small number of oxides is known to exhibit superconductivity, among them the Li-Ti-O system with onsets of superconductivity as high as 13,7 K, as reported by D.C. Johnston, H. Prakash, W.H. Zachariasen and R. Visvanathan in Mat. Res. Bull. 8 (1973) 777. Other known superconductive oxides include Nb-doped SrTiO<sub>3</sub> and BaPb<sub>1-q</sub>Bl<sub>q</sub>O<sub>3</sub>, reported respectively by A. Baratoff and G. Binnig in Physics 108B (1981) 1335, and by A.W. Sleight, J.L. Gillson and F.E. Bierstedt in Solid State Commun. 17 (1975) 27.

The X-ray analysis conducted Ly Johnston et al. revealed the presence in their Li-Ti-U system of three different crystallographic phases, one of them, with a spinel structure, showing the high critical temperature. The Ba-La-Cu-O system, too, exhibits a number of crystallographic phases, namely with mixed-valent copper constituents which have itinerant electronic states between non-Jahn-Teller Cu<sup>3</sup> and Jahn-Teller Cu<sup>2</sup> ions.

This applies likewise to systems where nickel is used in place of copper, with Ni<sup>3</sup> being the Jahn-Teller constituent, and Ni<sup>2</sup> being the non-Jahn-Teller constituent.

The existence of Jahn-Teller polarons is conducting crystals was postulated theoretically by K.H. Hoeck, H. Nickisch and H. Thomas in Helv. Phys. Acta 58 (1983) 237. Polarons have large electron-phonon interactions and, therefore, are favorable to the occurrance of superconductivity at high critical temperatures.

Generally, the Ba-La-Cu-O system, when subjected to X-ray analysis reveales three individual crystallographic phases, viz.

- a first layer-type perovskite-like phase, related to the K-NiFa structure, with the general composition Laz\_Ba\_CuO<sub>4++</sub>, with X≪1 and y≥0;
- a second, non-conducting CuO phase; and
- a third, nearly cubic perovskite phase of the general composition La<sub>1.x</sub>Ba<sub>x</sub>CuO<sub>3-y</sub> which appears to be independent of the exact starting composition.

as has been reported in the paper by J.G. Bednorz and K.A. Müller in Z. Phys. B - Condensed Matter 64 (1986) 189-193. Of these three phases the first one appears to be responsible for the high-T<sub>c</sub> superconductivity, the critical temperature showing a dependence on the barium concentration in that phase. Obviously, the Ba<sup>2</sup> substitution causes a mixed-valent state of Cu<sup>2</sup> and Cu<sup>3</sup> to preserve charge neutrality. It is assumed that the oxygen deficiency, y, is the same in the doped and undoped crystallites.

Both La<sub>2</sub>CuO<sub>4</sub> and LaCuO<sub>3</sub> are metallic conduc-

tors at high temperatures in the absence of barium. Actually, both are metals like LaNiO<sub>3</sub>. Despite their rinetallic character, the Ba-La-Cu-O type materials are ceramics, as are the other compounds of the RE<sub>2</sub>TM.O<sub>4</sub> type, and their manufacture more or less follows the known principles of ceramic fabrication. The preparation of a Ba-La-Cu-O compound, for example, in accordance with the present invention typically involves the following manufacturing steps:

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- Preparing aqueous solutions of the respective nitrates of barium, lanthanum and copper and coprecipitation thereof in their appropriate ratios.
- Adding the coprecipitate to oxalic acid and forming an intimate mixture of the respective oxalates.
- Decomposing the precipitate and causing a solidstate reaction by heating the precipitate to a temperature between 500 and 1200°C for one to eight hours.
- Pressing the resulting product at a pressure of about 4 kbar to form pellets.
- Re-heating the pellets to a temperature between 500 and 900°C for one half to three hours for sintering.

It will be evident to those skilled in the art that if the partial substitution of the lanthanum by strontium or calcium is desired, the particular nitrate thereof will have to be used in place of the barium nitrate of the example described above. Also, if the copper of this example is to be replaced by another transition metal, the nitrate thereof will obviously have to be employed.

Experiments have shown that the partial contents of the individual compounds in the starting composition play an important role in the formation of the phases present in the final product. While, as mentioned above, the final Ba-La-Cu-O system obtained generally contains the said three phases, with the second phase being present only to a very small amount, the partial substitution of lanthanum by strontium or calcium (and perhaps beryllium) will result in only one phase existing in the final  $La_{2-x}Sr_xCuO_{4-y}$  or  $La_{2-x}Ca_xCuO_{4-y}$ , respectively, provided x < 0.3.

With a ratio of 1:1 for the respective (Ba, La) and Cu contents, one may expect the said three phases to occur in the final product. Setting aside the said second phase, i.e. the CuO phase, whose amount is negligible, the relative volume amounts of the other two phases are depender, on the barium contents in the  $La_{2...}Ba_{n}CuO_{4...}$  complex. At the 1:1 ratio and with an x = 0.02, the onset of a localization transition is observed, i.e., the resistivity increases with decreasing temperature, and there is no superconductivity.

With x=0.1 at the same 1:1 ratio, there is a resistivity drop at the very high critical temperature of 35 K.

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With a (Ba,La) versus Cu ratio of 2:1 in the starting composition, the composition of the LarCuOaBa phase, which was assumed to be responsible for the serconductivity, is imitated, with the result that now only two phases are present, the CuO phase not existing. With a barium content of x=0.15, the resistivity drop occurs at  $T_c=26$  K.

The method for preparing the Ba-La-Cu-O complex involves two heat treatments for the precipitate at an elevated temperature for several hours. In the experiments carried out in connection with the present invention it was found that best results were obtained at 900°C for a decomposition and reaction period of 5 hours, and again at 900°C for a sintering period of one hour. These values apply to a ratio 1:1 composition as well as to a 2:1 composition.

For the ratio 2:1 composition, a somewhat higher temperature is permissible owing to the melting point of the composition being higher in the absence of excess copper oxide. Yet it is not possible by high-temperature treatment to obtain a one-phase compound.

Measurements of the dc conductivity were conducted between 300 and 4.2 K. For barium-doped samples, for example, with x < 0.3, at current densities of 0.5 A/cm<sup>2</sup>, a high-temperature metallic behavior with an increase in resistivity at low temperatures was found. At still lower temperatures, a sharp drop in resistivity (>90%) occurred which for higher current densities became partially suppressed. This characteristic drop was studied as a function of the annealing conditions, i.e. temperature and oxygen partial pressure. For samples annealed in air, the transition from itinerant to localized behavior was not found to be very pronounced, annealing in a slightly reducing atmosphere, however, led to an increase in resistivity and a more pronounced localization effect. At the same time, the onset of the resistivity drop was shifted towards higher values of the critical temperature. Longer annealing times, however, completely destroy the superconductivity.

Cooling the samples from room temperature, the resistivity data first show a metal-like decrease. At low temperatures, a change to an increase occurs in the case of Ca compounds and for the Basubstituted samples. This increase is followed by a resistivity drop, showing the onset of superconductivity at 22±2 K and 33±2 K for the Ca and Ba compounds, respectively. In the Sr compound, the resistivity remains metallic down to the resistivity drop at 40±1 K. The presence of localization effects, however, depends strongly on alkaline-earth ion concentration and sample preparation, that is to say, annealing conditions and also on the density which have to be optimized. All samples with low

concentrations of Ca. Sr. and Ba show a strong tendency:) localization before the resistivity drop occur.

Apparently, the onset of the superconductivity, i.e. the value of the critical temperature  $T_{\rm e}$ , is dependent, among other parameters, on the oxygen content of the final compound. It seems that a certain oxygen deficiency is necessary for the material to have a high- $T_{\rm e}$  behavior. In accordance with the present invention, the method described above for making the La<sub>2</sub>CuO<sub>4</sub>:Ba complex is complemented by an annealing step during which the oxygen content of the final product can be adjusted. Of course, what was said in connection with the formation of the La<sub>2</sub>CuO<sub>4</sub>:Ba compound, likewise applies to other compounds of the general formula RE<sub>2</sub>TM.O<sub>4</sub>:AE, such as, e.g. Nd<sub>2</sub>NiO<sub>4</sub>:Sr.

In the cases where a heat treatment for decomposition and reaction and/or for sintering was performed at a relatively low temperature, i.e. at no more than 950°C, the final product is subjected to an annealing step at about 900°C for about one hour in a reducing atmosphere. It is assumed that the net effect of this annealing step is a removal of oxygen atoms from certain locations in the matrix of the RE<sub>2</sub>TM.O<sub>4</sub> complex, thus creating a distortion in its crystalline structure. The O<sub>2</sub> partial pressure for annealing in this case may be between 10° and 10° bar.

In those cases where a relatively high temperature (i.e. above 950°C) was employed for the heat treatment, it might be advantageous to perform the annealing step in a slightly oxidizing atmosphere. This would make up for an assumed exaggerated removal of oxygen atoms from the system owing to the high temperature and resulting in a too severe distortion of the system's crystalline structure.

Resistivity and susceptibility measurements, as a function of temperature, of  $Sr^2$  and  $Ca^2$ -doped  $La_2CuO_{\Phi\gamma}$  ceramics show the same general tendency as the  $Ba^2$ -doped samples: A drop in resistivity  $\rho(T)$ , and a crossover to diamagnetism at a slightly lower temperature. The samples containing  $Sr^2$  actually yielded a higher onset than those containing  $Ba^2$  and  $Ca^2$ . Furthermore, the diamagnetic susceptibility is about three times as large as for the Ba samples. As the ionic radius of  $Sr^2$  nearly matches the one of  $La^3$ , it seems that the size effect does not cause the occurrence of superconductivity. On the contrary, it is rather adverse, as the data on  $Ba^2$  and  $Ca^2$  indicate.

The highest  $T_c$ 's for each of the dopant ions investigated occur for those concentrations where, at room temperature, the  $Re_{2,u}TM_vO_{4,v}$  structure is close to the orthorhombic-tstragonal structural phase transition which may be related to the substantial electron-phonon interaction enhanced by the substitution. The alkaline-earth substitution of

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the rare earth metal is clearly important, and quite likely creates TM ions with no  $e_0$  Jahn-Teller orbitals. Therefore, the absence of these J.-T. orbitals, that is, J.-T. holes near the Fermi energy probably plays an important role for the  $T_e$  enhancement.

#### Claims

- 1) Superconductive compound of the RE<sub>2</sub>TM.O<sub>4</sub> type having a transition temperature above 26 K, wherein the rare earth (RE) is partially substituted by one or more members of the alkaline earth groups of elements (AE), and wherein the oxygen content is adjusted such that the resulting crystal structure is distorted and comprises a phase of the general composition RE<sub>2.a</sub>AE<sub>a</sub>TM.O<sub>4.y</sub>, wherein TM represents a transition metal, and x < 0.3 and y < 0.5.
- 2) Compound in accordance with claim 1, wherein the rare earth (RE) is lanthanum and the transition metal (TM) is copper.
- Compound in accordance with claim 1, wherein the rare earth is cerium and the transition metal is nickel.
- 4) Compound in accordance with claim 1, wherein the rare earth is lanthanum and the transition metal is nickel.
- 5) Compound in accordance with claim 1, wherein barium is used as a partial substitute for the rare earth, with x < 0.3 and  $0.1 \le y \le 0.5$ .
- 6) Compound in accordance with claim 1, wherein calcium is used as a partial substitute for the rare earth, with x < 0.3 and  $0.1 \le y \le 0.5$ .
- 7) Compound in accordance with claim 1, wherein strontium is used as a partial substitute for the rare earth, with x < 0.3 and  $0.1 \le y \le 0.5$ .
- 8) Compound in accordance with claim 1, wherein the rare earth is lanthanum and the transition metal is chromium.
- Compound is accordance with claim 1, wherein the rare earth is neodymium and the transition metal is copper.
- 10) Method for making superconductive compounds of the RE<sub>2</sub>TM.O<sub>4</sub> type, with RE being a rare earth, TM being a transition metal, the compounds having a transition temperature above 28 K, comprising the steps of:
- preparing aqueous solutions of the nitrates of the rare earth and transition metal constituents and of one or more of the alkaline earth metals and coprecipitation thereof in their appropriate ratios;
- adding the coprecipitate to oxalic acid and forming an intimate mixture of the respective oxalates:
   decomposing the precipitate and causing a solid-state reaction by heating the precipitate to a temperature between 500 and 1200°C for a period of

time between one and eight hours:

- allowing the resultant powder product to cool;
- pressing the powder at a pressure of between 2 and 10 kbar to form pellets;
- re-adjusting the temperature of the pellets to a value between 500 and 1000°C for a period of time between one half and three hours for sintering;
- subjecting the pellets to an additional annealing treatment at a temperature between 500 and 1200°C for a period of time between one half and 5 hours in a protected atmosphere permitting the adjustment of the oxygen content of the final product which has a final composition of the form RE<sub>2</sub>.  $_{\rm a}$ TM.O<sub>4y</sub>, wherein x < 0.3 and 0.1 < y < 0.5.
- 11) Method in accordance with claim 10, wherein the protected atmosphere is pure oxygen.
- 12) Method in accordance with claim 10, wherein the protected almosphere is a reducing atmosphere with an oxygen partial pressure between 10 <sup>1</sup> and 10 <sup>5</sup> bar.
- 13) Method in accordance with claim 10, wherein the decomposition step is performed at a temperature of 900°C for 5 hours, and wherein the annealing step is performed at a temperature of 900°C for one hour in a reducing atmosphere with an oxygen partial pressure between 10 1 and 10 3 par.
- 14) Method in accordance with claim 10, wherein lanthanum is used as the rare earth and copper is used as the transition metal, and wherein barium is used to partially substitute for the lanthanum, with x < 0.2, wherein the decomposition step is performed at a temperature of 900°C for 5 hours, and wherein the annealing step is performed in a reducing atmosphere with an oxygen partial pressure on the order of 10  $^{3}$  bar and at a temperature of 900°C for one hour.
- 15) Method in accordance with claim 10, wherein lanthanum is used as the rare earth and nickel is used as the transition metal, and wherein barlum is used to partially substitute for the lanthanum, with x < 0.2, wherein the decomposition step is performed at a temperature of 900°C for 5 hours, and wherein the annealing step is performed in a reducing atmosphere with an oxygen partial pressure on the order of  $10^{-3}$  bar and at a temperature of 900°C for one hour.
- 16) Method in accordance with claim 10, wherein lanthanum is used as the rare earth and copper is used as the transition metal, and wherein caicium is used to partially substitute for the lanthanum, with x < 0.2, wherein the decomposition step is performed at a temperature of 900°C for 5 hours, and wherein the annealing step is performed in a reducing atmosphere with an oxygen partial pressure on the order of 10 3 bar and at a temperature of 900°C for one hour.

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17) Method in accordance with claim 10, wherein fanthanum is used as the rare earth and copper is used as the transition metal, and wherein strontium is used to partially substitute for the lanthanum, with x < 0.2, wherein the decomposition step is performed at a temperature of 900°C for 5 hours, and wherein the annealing step is performed in a reducing atmosphere with an oxygen partial pressure on the order of  $10^{-3}$  bar and at a temperature of  $900^{\circ}$ C for one hour.

18) Method in accordance with claim 10, wherein cerium is used as the rare earth and nickel is used as the transition metal, and wherein barlum is used to partially substitute for the cerium, with x < 0.2, wherein the decomposition step is performed at a temperature of 900°C for 5 hours, and wherein the annealing step is performed in a reducing atmosphere with an oxygen partial pressure on the order of 10  $^3$  bar and at a temperature of 900°C for one hour.



### **EUROPEAN SEARCH REPORT**

Application number

EP 87 10 0961

Category		ith indication, where appropriate,	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. CI 4)
•	REVUE DE CHIMIE 21, 1984, pages FR; C. MICHEL et intercalation in copper oxides re perovskites" * page 417, p	407-425, Paris, al.: "Oxygen mixed valence	1	H O1 L 39/12
	425 * 			
	·			TECHNICAL FIELDS SEARCHED (Int. CI.4)
				H 01 L 39/00
				·
	The present search report has t	een drawn up for all claims		
Place of search BERLIN		Date of completion of the search 17-07-1987	ROUSS	Exeminer SEL A T
Y : pa	CATEGORY OF CITED DOCL rticularly relevant if taken alone rticularly relevant if combined w current of the same category chnological background	E : earlier pa after the f ith another D : documen	tent document,	



#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J.G. Bednorz et al.

Date: April 11, 1996

Serial No.: 08/303,561

Group Art Unit: 1105

Filed: September 9, 1994

Examiner: D. McGinty

For: SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION

TEMPERATURE, AND METHODS FOR THEIR USE AND PREPARATION

I hereby certify that this paper is being facsimile transmitted under Rule 37 CFR § 1.161(d) to the U.S. Patent and Trademark Office on the date shown above.

Daniel P. Morris

Reg. No. 32,053

The Commissioner of Patents and Trademarks Washington, D.C. 20231

### **SUPPLEMENTARY RESPONSE**

Sir:

In response to the Office Action dated March 19, 1995, please consider the following:

#### **REMARKS**

These remarks are in addition to those of the previously submitted response.

Claims 24-26, 86-90 and 96-108 have been rejected under 35 USC § 102(a) as being anticipated by the Asahi Shinbum article and under 35 USC § 103 in view of the Asahi

Shinbum article. In addition to Applicants' remarks in regard to this rejection in Applicant's prior response, please consider the following:

The date of the Asahi Shinbum article is November 28, 1986. As stated in Applicants' specification at page 6, lines 7-10:

The basis for our invention has been described by us in the following previously published article: J.G. Bednorz and K.A. Mueller, Zeitschrift for Physik B - Condensed Matter, 64, pp. 189-193, Sept. (1986).

The Examiner is using Asahi Shinbum as a reference under 35 USC § 102(a). Applicants respectfully disagree since to do so does not permit Applicants the one year period provided under 35 USC § 102(b) to file a U.S. application after their own publication which permitted Applicants to file the present application up to September 1987. The date of the Asahi Shinbum article is after the date of Applicants' publication.

In regard to the two-year grace period under a prior statute, the U.S. Supreme Court in Andrews v. Hovey, 123 US 267 (1887) states that:

"The evident purpose of the section was to fix a period of limitation which should be certain, and require only a calculation of time, and should not depend upon the uncertain question of whether the Applicant had consented to or allowed the sale or use. Its object was to require the inventor to see to it that he filed his application within two years from the completion of his invention, so as to cut off

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all question of the defeat of his patent by a use or sale of it by others more than two years prior to his application, and thus leave open only the question of priority of invention. The evident intention of congress was to take away the right (which existed under the act of 1836) to obtain a patent after an invention had for a long period of time been in public use, without the consent or allowance of the inventor; it limited that period to two years, whether the inventor had or had not consented to or allowed the public use."

From this quote from Andrews v. Hovey, it is evident that the use or sale by others prior to filing a patent application by the inventor does not cut off the inventors right to obtain a patent so long as the inventor files the application within the statutory period which was 2 years at the time of the Andrews v. Hovey decision and is now 1 year under 35 USC 102(b).

The Patent Office Board of Appeals in Ex parte Powell and Davies, 37 USPQ 285 states in regard to the publication of Applicants foreign patent application before the filing of a U.S. application on October 5, 1936 on an invention described in the foreign patent application that:

The Examiner has also rejected the claims on the printed specification of Applicants' own British application which appears from this record to have been published on August 27, 1936. We know of no authority for such a rejection. Neither section 3886 nor section 4887 R.S. warrants the rejection. Obviously, the publication could not have a date

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prior to Applicants' invention. There is no statute that requires an Applicant to make his invention in this country.

Therefore, Applicants of the present invention can rely on their publication in Zeitschrift for Physik as evidence of their invention.

The Patent Office Board of Appeals in Ex parte Powell and Davies, 37 USPQ 285, 286 further states:

The Commissioner indicates in Ex parte Grosselin that the Examiner should consider whether the German patent was derived from Applicant and was in effect nothing more than a printed publication of Grosselin's invention.

The Asahi Shinbum article states in the first paragraph:

A new ceramic with a very high  $T_c$  of 30K of the superconducting transition has been found. The possibility of high  $T_c$  - superconductivity has been reported by scientists in Switzerland this spring. The group of Prof. Shoji TANAKA, Dept. Appl. Phys. Faculty of Engineering at the University of Tokyo confirmed in November, that this is true.

The "scientists in Switzerland" are the inventors of the above-identified application. The Asahi Shinbum article only reports the work of Applicants and that it was reproduced by Prof. Tanaka. This article is a disclosure of Applicants' "own invention" and clearly

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in the words of the Board in Ex parte Powell and Davies, "was derived from [Applicants] and [is] in effect nothing more than a printed publication of [Applicants'] own invention and cannot be used as a reference.

The Patent Office Board of Appeals in Ex parte Lemieux 148, 140 states that:

Finally, we believe that our holding is consistent with decisions in interference practice wherein, even though in the usual case a party may not establish a priority date of invention by reference to activity in a foreign country, yet in an originality case where a party is seeking to prove that the other party derived from him so that there is only a single original inventor, he may be permitted to prove derivation by reference to activity abroad. ... By analogy, in the present case appellant has demonstrated that he is the single original inventor, there being no adverse party.

Following this decision it is clear from the Asahi Shinbum article that Applicants are the "single original inventor" and that the Asahi Shinbum article is "derived" from Applicants and that Professor Tanaka's work reported in the Asahi Shinbum article is "derived" from Applicants.

Therefore, the Examiner is respectfully requested to withdraw the rejection of claims 24-26, 86-90 and 96-108 under 35 USC § 102(a) as anticipated by Asahi Shinbum and under 35 USC § 103 as obvious over Asahi Shinbum.

Attached are copies of the following decisions:

Ex parte Powell and Davies 37 USPQ 285 Ex parte Lemieux 115 USPQ 148

Respectfully submitted,

Daniel P. Morris

Reg. No. 32,053

IBM Corporation Intellectual Property Law Dept. P.O. Box 218 Yorktown Heights, N.Y. 10598 (914) 945-3217

Patent Office Board of Appeals

Ex parte LEMIEUX
Patent issued Oct. 8, 1957
Opinion dated July 31, 1957

#### PATENTS

1. Interference—Reduction to practice— In general (§ 41.751)

Patentability—Anticipation—In general (§ 51.201)

Patentability — Anticipation — Publications—In general (§ 51.2271)

Act of August 8, 1946 (35 U.S.C. 104) was enacted to overrule Electric v. Shimadzu, 307 U.S. 5, 41 USPQ 155, and preclude applicant or patentee from relying upon foreign activity to establish date of invention; it had no effect on Exparte Powell, 37 USPQ 285; hence, inventor's foreign publication within year prior to filing United States application does not bar him from obtaining patent.

2. Affidavits — Anticipating references (§ 12.3)

Rule 131 does not apply where publication is publication of applicant's own invention; domestic inventors are not distinguished from foreign inventors; all that is required is that identity of application inventor and publication author be established.

3. Interference — Originality of invention—In general (§ 41.551)

Interference—Reduction to practice— In general (§ 41.751)

Even though in usual case interference party may not establish priority date of invention by reference to activity in foreign country, in originality case, where party is seeking to prove that opponent derived from him so that there is only a single original inventor, he may be permitted to prove derivation by reference to activity abroad.

Particular patents—Ustilic Acids 2,809,205, Lemieux, Production of Ustilic Acids, claims 1 to 4 and 6 of application allowed.

Appeal from Division 63.
Application for patent of Raymond U.
Lemieux, Serial No. 281,451, filed Apr.
9, 1952. From decision rejecting claims 1
to 4 and 6, applicant appeals. Reversed.
PIERCE, SCHEFFLER & PARKER, Washington, D.C., and ALEX E. MACRAE for applicant.

Before DUNCOMBE, Examiner in Chief, and MAGIL and BREWRINK, Acting Examiners in Chief.

MAGIL, Acting Examiner in Chief.

This is an appeal from the final rejection of claims 1 through 4 and 6. Claims 5 and 7, the remaining claims in the case, have been withdrawn from further consideration in accordance with Rule 142(b) and are not before us.

Since the issue involved in this case is purely legal in nature, there is no reason for reproducing an illustrative claim.

The reference relied upon is:

Lemieux, Canadian Journal of Chemistry, Vol. 29, (May 1951), pages 415-425.

We need not refer to the subject matter of the claims because, as previously indicated, the appeal involves only a legal point. The following facts are not in dispute:

1. The appellant is the author of the cited publication.

2. The subject matter of the appealed claims is adequately disclosed in the cited publication.

3. The cited article was published prior to appellant's filing date in this country, but not more than one year prior thereto.

4. Appellant does not rely upon any earlier filing date to antedate the publication, nor does he assert that he completed the invention in this country prior to the date of the publication.

The examiner holds that appellant is barred from obtaining a patent by the provision of 35 U.S.C. 102(a) and that he cannot avoid this bar because of the restriction of 35 U.S.C. 104 and the words "in this country" in Rule 131.

Appellant contends that he filed his application within the one year period specified in 35 U.S.C. 102(b) and that Rule 131 is inapplicable. Appellant relies upon the case of Ex parte Powell and Davies, 489 O.G. 231, 1938 C.D. 15, 37 USPQ 285; he also refers to the International Convention for the Protection of Industrial Property and to the effect of the examiner's rejection on Canadian inventors.

We have carefully considered the examiner's rejection and the appellant's arguments and have studied the pertiment cases on this subject. On its face, and disregarding subsequent statutory changes, the Powell and Davies case appears to be most apposite. The examiner recognizes that the cited decision is relevant, but holds that it is no longer controlling because of the Act of August 8, 1946, which resulted in the enactment of the statute presently corresponding to 35 U.S.C. 104.

[1] We hav U.S.C. 104 and by the case o Shimadzu et a 675, 83 Law. 504 O.G. 4, 41 in an infringer not precluded invention by abroad. We r the Shimadzu (country" in ol to present Rul the case of In 810, 1942 C.D F.2d 169, 52 U the Shimadzu anomalous situ plying in inte another rule a ings. In order the Act of Au and, as is evi No. 1502, June 2nd Session, ar January 28, Session, the p effect, to overr tation of the S clude an appl

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[1] We have traced the history of 35 U.S.C. 104 and find that it was prompted by the case of Electric Storage Co. v. Shimadzu et al., 307 U.S. 5, 59 Sup.Ct. 675, 83 Law. Ed. 1071, 1939 C.D. 870, 504 O.G. 4, 41 USPQ 155, which held that in an infringement action the patentee is not precluded from proving his date of invention by reference to his activity abroad. We note that, on the basis of the Shimadzu decision, the words "in this country" in old Rule 75, corresponding to present Rule 131, were held invalid in the case of In re McFarlane, 29 C.C.P.A. 810, 1942 C.D. 254, 540 O.G. 237, 125 F.2d 169, 52 USPQ 335. The decision in the Shimadzu et al. case created an anomalous situation, with one rule applying in interference proceedings and another rule applying in other proceedings. In order to remedy this situation, the Act of August 8, 1946 was enacted and, as is evident from Senate Report No. 1502, June 14, 1946, 79th Congress, 2nd Session, and House Report No. 1498, January 28, 1946, 79th Congress, 2nd Session, the purpose of the law was, in effect, to overrule the statutory interpretation of the Shimadzu decision and preclude an applicant or a patentee from relying upon foreign activity to establish a date of invention. The Shimadzu et al. case is referred to by name in both the Senate and the House Reports.

With the foregoing analysis, it may be said that the purpose and effect of the Act of August 8, 1946 was solely to overrule the Shimadzu et al. case and it had no effect upon the Powell and Davies decision cited by appellant. The Powell and Davies decision being unaffected, we should hold that it is controlling in the present case and dictates reversal of the examiner's decision. note that the Powell and Davies case was cited by the appellant in the case of In re Saurer, 28 C.C.P.A. 1021, 1941 C.D. 405, 529 O.G. 802, 118 F.2d 719, 49 USPQ 78, but the Court found the decision in-applicable only because the appellant failed to establish his identity with the person named in the reference. We may also state that we consider the case of Ex parte Grosselin, 97 O.G. 2977, 1901 C.D. 248, cited in the Powell and Davies decision, as well as the earlier case of Ex parte Grosselin, 84 O.G. 1284, 1898 C.D. 163, to be pertinent.

Aside from the history of 35 U.S.C. 104, there is another reason why this section is not applicable to the present case. Appellant is not seeking to "establish a date of invention" but has merely argued that he is the author of the cited publication and this is not disputed by the examiner. There being no evidence of invention by anyone else prior to

appellant's filing date, the date of appellant's invention is immaterial. In the present case, we are not concerned with appellant's "date of invention" visavis the publication of another, an interferant, or other adverse party.

[2] Insofar as the requirements of Rule 131 are concerned, we need do nothing more than refer to and state our agreement with the holdings in the first Grosselin decision (84 O.G. 1284, 1898 C.D. 163) and in the Powell and Davies case concerning old Rule 75, corresponding to present Rule 131. In the former decision it was held that "this rule presupposes that the printed publication is the publication by some one other than the applicant whose application is rejected-by some one who asserts inventorship therein either in himself or some other person than the applicant." In the later case it was held that the rule is not "intended to apply to a case where the publication appears without question to be a publication of the applicant's own invention."

We also take cognizance of several decisions (Ex parte Ensign, 2 USPQ 214; Ex parte Layne, 63 USPQ 17; Ex parte Hirschler, 110 USPQ 384) which have held that, apparently in the case of a domestic inventor, a publication dated less than a year prior to the filing date of an application is not an effective bar if the applicant makes a satisfactory showing that the publication is his own invention or that he is, in effect, the author of the publication. In none of these cases is there any indication that the applicant made the usual showing under Rule 131, that is, reduction to practice prior to the date of the publication or conception prior to the date of the publication coupled with the necessary diligence. We find no reason for distinguishing between a domestic inventor and a foreign inventor in situations of this type and all that is required is that the identity of the application inventor and the publication author be established. There is no dispute on this point in the present case.

[8] Finally, we believe that our holding is consistent with decisions in interference practice wherein, even though in the usual case a party may not establish a priority date of invention by reference to activity in a foreign country, yet in an originality case where a party is seeking to prove that the other party derived from him so that there is only a single original inventor, he may be permitted to prove derivation by reference to activity abroad. Shiels v. Lawrence and Kennedy, 81 O.G. 2085, 1897 C.D. 184; Stiff v. Galbraith, 108 O.G. 290, 1904 C.D. 10. By analogy, in the present case appellant has demonstrated

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that he is the single original inventor, there being no adverse party.

In accordance with the foregoing reasoning, we conclude that the examiner's rejection cannot be sustained. We do not consider it necessary to discuss appellant's arguments concerning the International Convention for the Protection of Industrial Property or the effect of the examiner's rejection on Canadian inventors.

The decision of the examiner is reversed.

Patent Office Board of Appeals

Ex parte BERGMANN

Patent issued Oct. 8, 1957

Opinion dated Jan. 22, 1957

#### PATENTS

1. Patentability — Change — Material (§ 51.257)

Claims are allowed where applicant did not merely indulge in routine experimentation with material having properties which would be expected to produce results desired, but utilized to advantage a material whose properties appeared to be unfavorable.

Particular patents—Sound Recording 2,809,237, Bergmann, Magnetic Sound Recording Head, claims 1 and 3 to 7 of application allowed.

Appeal from Division 16.
Application for patent of Friedrich
Bergmann, Serial No. 209,250, filed Feb.
3, 1951. From decision rejecting claims
1 and 3 to 7, applicant appeals. Reversed.
MARZALL, JOHNSTON, COOK & ROOT,
Chicago, Ill., for applicant.

Before TAYLOR and KREEK, Examiners in Chief, and NILSON, Acting Exminer in Chief.

KREEK, Examiner in Chief.

This is an appeal from the final rejection of claims 1 and 3 to 7 inclusive. No claims have been allowed.

Claim 1 is illustrative:

1. Magnetic sound recording head having a core consisting exclusively of magnetic ferrite.

The references relied on are:

Burns 2,536,260 Jan. 2,1951 Buhrendorf 2,592,652 Apr. 15, 1952

As is apparent from the illustrative claim, the subject matter here on appeal relates to a recording head for a magnetic sound recorder in which the core consists exclusively of magnetic ferrite. Numerous advantages are claimed for this construction among which are reduction of wear on the core as a result of the magnetic record medium passing thereover, as compared to the wear of conventional iron cores under similar circumstances; and reduction in electrical losses especially at high frequencies.

Claims 1 and 3 to 7 were rejected as being unpatentable over Buhrendorf or Burns in view of the general knowledge of the art, the examiner's position being "The routine examination of any known substance for a particular use is expected where the known basic requirements of the use are compatible with some characteristics of the substance." It is his opinion "that the mere knowledge that 'ferrites' are magnetic is enough to warrant investigation by workers in magnetic recording. The knowledge of their high frequency losses and avowed utility in electo-acoustic devices practically demands investigation."

Appellant contends that the references relied on do not suggest making cores exclusively of ferrite, and that the known permeability, saturation and abrasive characteristics of ferrite would point away from its use in sound recording heads rather than suggest it. He asserts that recording heads heretofore used are made with cores of highly permeable material to secure proper operation, but that satisfactory operation is secured with ferrite cores even though the permeability thereof is considerably less than the magnetic materials previously used He asserts that the smaller magnetic saturation of ferrite as compared with metallic magnetic material would tend to indicate its unsuitability in erasing heads where high magnetic saturation is required He further asserts that the fact ferrite would not abrade the surface of the sound band was surprising since sintered ferrite behaves somewhat like sintered porcelain. This characterists which would have been expected to be det rimental is alleged to provide a great advantage resulting in heads having life at least ten times longer than that of metal heads heretofore used. Appel lant has made of record a publication by Rolf Cruel in Technische Hausmittellung gen des Nordwestdeutschen Rundfunk which compares magnetic heads made with ferrite with previously used laminated, high permeability iron alloy, which publication demonstrates important technical advantages possessed by ferrit over previously used magnetic material These are summarized as greater hard

ness resulting in gr sistance to wear so used for much longwithout adjustment, high frequency and ? for erasing.

We have carefully in view of appellant' various publications show the suitability material for recording of which we are of the rejection cannot be so

The patent to Bu magnetic recording h netic materials such a ing high permeability. tions are made extre 0.001 of an inch thic reduce eddy current? value as is feasible c ability to work with tions. Obviously Bu anticipatory value, but the problems confronthis field as of the tin Buhrendorf application

Burns shows a magn for magnetic record utilize a central yoke either side of which : fron pole pieces 11 and contact the surface of used in the recorder. forms a part of the cir tor, the frequency of a result of the variatio flow through the ferr noted, however, that t pieces are necessary to netic tape from being frequency currents flo wound in the ferrite co no suggestion in Burr core may be made of f [1] The examiner a

that low permeability factor as he states "su be undesirable permeal undesigned results." I the teachings of the point away from the material having low I core for a sound recor withstanding this, th clearly shows that in s permeability of ferrite, actic properties are s forms satisfactorily as a magnetic recording alficantly, however, is th made of ferrite have 1 resistance to abrasion v operate up to ten times ventional metallic iron Without adjustment. important factor in rac

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Patent Office Board of Appeals Ex parte Powell and Davies

Appl. No. 23985

Patent issued Apr. 5, 1938-Opinion dated Mar. 1, 1938

Patents-Patentability-Anticipation-Foreign patents; Affidavits-Anticipating ref-

Applicants' own British patent has not been sealed, so there is no reason for registry under Rule 29; the British application was published in Aug., 1936, but there is no authority for basing on it rejection of United States application filed less than two years after such publication; Rule 75 was intended to provide ex parte means by which applicant can overcome rejection based on publication not more than two years arise to his invention but described by the provider than two years arise to his invention but described by the provider to his invention between the historia between the provider to his invention between the historia betwee than two years prior to his invention but does not apply to case where publication appears without question to be publication of applicant's own invention.

Patents—Electrodeposition of Silver— 2113517, Powell and Davies, Electrodeposition of Silver, claims 1, 2, 4 to 7, 10 and 11 of application allowed.

Patent No. 2113517 for electrodeposition of silver issued on application filed Oct. 5, 1936.

Appeal from Division 56.

Howson & Howson for applicants.

Before VAN ARSDALE, Assistant Commissioner, and REDROW and PORTER, Examiners in Chief.

PORTER, Examiner in Chief .- This is an appeal from the final rejection of claims 1, 2, 4 to 7, 10 and 11.

Claim 7 is illustrative.

7. A plating bath comprising a potassium argento-cyanide, an excess of free potassium cyanide, carbon disulphide and Turkey red oil.

The references relied upon are as fol-

Schlotter (British) 443,428, Feb. 27, 1936.

Powell et al (British) 450,979, Aug.

27, 1936. Blum & Hogaboom, Principles of Electroplating (2nd Ed.) 1930, pages 350, 355 to 357.

It appears from Blum and Hogaboom that the silver plating solution of the claims is old except for the addition of alkali metal soaps or their equivalents. Blum et al describe an excess of free alkali metal cyanide but the British patent indicates that this excess should be very large and describes the use of Turkey red oil to which applicants refer at the bottom of page 3 of their specification. The British patent does not suggest the use of carbon bisulphide but rather suggests a substitute therefor. Blum et al does not suggest the use of soap. It is the combined use of carbon bisulphide and soap in the silver plating which applicants describe as their invention. The gist of the examiner's position with respect to the references referred

to appears to be that there is no invention in the combined use of the carbon bisulphide and soap in the silver plating bath. The trouble with this position is that it is not warranted by the record which does not show carbon bisulphide combined with a dispersing agent of the nature of the one employed by appli-

The examiner has also rejected the claims on the printed specification of applicants' own British application which appears from this record to have been published on August 27, 1936. We know of no authority for such a rejection. Neither section 4886 nor section 4887 R. S. warrants the rejection. Obviously, the publication could not have a date prior to applicants' invention. There is no statute that requires an applicant to make his invention in this country.

It does not appear that the British patent has been sealed which sealing would be necessary in the case of a British patent in a rejection under Rule 29 and it appears from the decision of the Supervisory Examiner (Paper No. 7) that the examiner's real position is not that applicants are barred by the provisions of Rule 29 as appears from his statement, but that applicants have failed to overcome their own publication by affidavits filed under Rule 75.

Applicants filed a petition to the Commissioner asking that the examiner be instructed to withdraw the citation of their own British specification as a ref-erence against the claims. This the Commissioner refused to do indicating that an adverse decision on the point by him might act to preclude a favorable decision by the Board of Appeals.

The examiner holds the affidavits insufficient as the nature of the contents of the notes referred to in the affidavit of Coussmaker does not appear and there is no such showing as to facts as is necessary in affidavits filed under Rule 75. The affidavits have been reexamined but we find nothing therein except the mere inference that the subject matter in issue here was disclosed to Stones by Coussmaker.

The case of Ex parte Grosselin 1901 C. D. 248, is analyzed by applicants in such a way as to contend that this decision never was intended to apply to a case where the printed publication in question was cne's own publication There are, however, certain obiter statements made in the Grosselin decision which might be taken to indicate that the provisions of Rule 75 requiring the applicants to show completion of the invention in this country apply to a case wherein the applicant is required to overcome the filing date of his own publication. It is our opinion, however, that these obiter statements are not definite and any such construction of Rule 75 as contended for by the examiner is clearly refuted by the general tenor and intent of the decision. Rule 75 was intended to provide Ex parte means by which an applicant can overcome a rejection based on a publication of the invention not more than two years prior to his application. We do not agree with the examiner that this rule is intended to apply to a case where the publication

appears without question to be a publication of the applicant's own invention.

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The Commissioner indicates in Ex parte Grosselin that the examiner should consider whether the German patent was derived from applicant and was in effect nothing more than a printed publication of Grosselin's invention. The decision further indicates that Rule 75 permits an applicant to make an ex parte showing of his rights (page 254) and that the whole proceeding is by analogy to the interference practice. On page 253 it is stated that, "Whenever this Office has satisfactory evidence that some other person is as against the applicant entitled to a patent, it is, under the general principles of the law which are well recognized, bound to reject the applica-

Applicants' patent in Great Britain has not been sealed so that there is no reason for rejecting the claims under Rule 29 and it appears obvious that applicants made their invention prior to the date of their published specification.

It is our opinion that this record does not show prior invention of the subject matter of the claims by a party other than the applicants. They are, therethan the applicants. fore, entitled to a patent.

The decision of the examiner is re-

versed.

Circuit Court of Appeals, Second Circuit SHELDON et al. (complainants-appellees)

MOREDALL REALTY CORPORATION (respondent-appellant) Decided Feb. 21, 1938 No. 139

Copyrights-Pleading and practice in courts; Appeals to Circuit Courts of Appeals-Orders appealable

Trial judge, recognizing non-existence of actual or threatened continued infringement, concluded in opinion that injunction should not issue, but apparently through inadvertence injunction was included in decree; appeal from that part of decree was properly taken; injunction is vacated and decree to that extent reversed; as jurisdiction to review interlocutory decree depends on 28 U. S. C. 227, general rule is that propriety of granting other relief forms no part of subject matter of appeal and is not before Circuit Court of Appeals, not being final decree, but rule is subject to one exception; where such appeal is rightly taken court may examine record thus made to determine whether bill is wholly lacking in equity and, if so, may dismiss; but where doubt exists as to equitable jurisdiction, that matter is left to appeal from final decree.

Copyrights-Pleading and practice in courts-Copyright statute differs from patent and trade mark statutes, and injunction is not condition precedent for accounting and award of damages for copyright infringement; equitable jurisdiction having been invoked in good faith by suitable allegations in bill, jurisdiction may not fall with failure of proof on merits of exclusively equitable rights; nor can court be sure on appeal from interlocutory decree that equity does not have consurrent invisition of accounting and principles equity does not have concurrent jurisdiction of accounting on general principles.

Patents-Jurisdiction of courts-For patent infringement-In patent cases, only where injunction is rightly granted may there be accounting and award of damages in equity.

# Possible High $T_c$ Superconductivity in the Ba-La-Cu-O System

J.G. Bednorz and K.A. Müller IBM Zürich Research Laboratory, Rüschlikon, Switzerland

Received April 17, 1986

Metallic, oxygen-deficient compounds in the Ba-La-Cu-O system, with the composition  $Ba_xLa_{5-x}Cu_5O_{5(3-y)}$  have been prepared in polycrystalline form. Samples with x=1 and 0.75, y>0, annealed below 900 °C under reducing conditions, consist of three phases, one of them a perovskite-like mixed-valent copper compound. Upon cooling, the samples show a linear decrease in resistivity, then an approximately logarithmic increase, interpreted as a beginning of localization. Finally an abrupt decrease by up to three orders of magnitude occurs, reminiscent of the onset of percolative superconductivity. The highest onset temperature is observed in the 30 K range. It is markedly reduced by high current densities. Thus, it results partially from the percolative nature, bute possibly also from 2D superconducting fluctuations of double perovskite layers of one of the phases present.

#### I. Introduction

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"At the extreme forefront of research in superconductivity is the empirical search for new materials" [1]. Transition-metal alloy compounds of A 15 (Nb<sub>3</sub>Sn) and B 1 (NbN) structure have so far shown the highest superconducting transition temperatures. Among many A 15 compounds, careful optimization of Nb—Ge thin films near the stoichiometric composition of Nb<sub>3</sub>Ge by Gavalev et al. and Testardi et al. a decade ago allowed them to reach the highest  $T_c$ = 23.3 K reported until now [2, 3]. The heavy Fermion systems with low Fermi energy, newly discovered, are not expected to reach very high  $T_c$ 's [4].

Only a small number of oxides is known to exhibit superconductivity. High-temperature superconductivity in the Li-Ti-O system with onsets as high as 13.7 K was reported by Johnston et al. [5]. Their x-ray analysis revealed the presence of three different crystallographic phases, one of them, with a spinel structure, showing the high  $T_c$  [5]. Other oxides like perovskites exhibit superconductivity despite their small carrier concentrations, n. In Nb-doped SrTiO<sub>3</sub>, with  $n = 2 \times 10^{20}$  cm<sup>-3</sup>, the plasma edge is below the highest optical phonon, which is therefore unshielded

[6]. This large electron-phonon coupling allows a  $T_c$ of 0.7 K [7] with Cooper pairing. The occurrence of high electron-phonon coupling in another metallic oxide, also a perovskite, became evident with the discovery of superconductivity in the mixed-valent compound BaPb<sub>1-x</sub>Bi<sub>x</sub>O<sub>3</sub> by Sleight et al., also a decade ago [8]. The highest  $T_c$  in homogeneous oxygen-deficient mixed crystals is 13 K with a comparatively low concentration of carries  $n = 2-4 \times 10^{21}$  cm<sup>-3</sup> [9]. Flat electronic bands and a strong breathing mode with a phonon feature near 100 cm<sup>-1</sup>, whose intensity is proportional to  $T_c$ , exist [10]. This last example indicates that within the BCS mechanism, one may find still higher Tc's in perovskite-type or related metallic oxides, if the electron-phonon interactions and the carrier densities at the Fermi level can be enhanced further.

Strong electron-phonon interactions in oxides can occur owing to polaron formation as well as in mixed-valent systems. A superconductivity (metallic) to bipolaronic (insulator) transition phase diagram was proposed theoretically by Chakraverty [11]. A mechanism for polaron formation is the Jahn-Teller effect, as studied by Höck et al. [12]. Isolated Fe<sup>4+</sup>, Ni<sup>3+</sup> and Cu<sup>2+</sup> in octahedral oxygen environment

show strong Jahn-Teller (J.T.) effects [13]. While SrFe(VI)O<sub>3</sub> is distorted perovskite insulator, LaNi(III)O<sub>3</sub> is a J.T. undistorted metal in which the transfer energy  $b_{\pi}$  of the J.T.  $e_{\pi}$  electrons is sufficiently large [14] to quench the J.T. distortion. In analogy to Chakraverty's phase diagram, a J.T.-type polaron formation may therefore be expected at the borderline of the metal-insulator transition in mixed perovskites, a subject on which we have recently carried out a series of investigations [15]. Here, we report on the synthesis and electrical measurements of compounds within the Ba-La-Cu-O system. This system exhibits a number of oxygen-deficient phases with mixed-valent copper constituents [16], i.e., with itinerant electronic states between the non-J.T. Cu<sup>3+</sup> and the J.T. Cu2+ ions, and thus was expected to have considerable electron-phonon coupling and metallic conductivity.

#### II. Experimental

#### 1. Sample Preparation and Characterization

Samples were prepared by a coprecipitation method from aqueous solutions [17] of Ba-, La- and Cu-nitrate (SPECPURE JMC) in their appropriate ratios. When added to an aqueous solution of oxalic acid as the precipitant, an intimate mixture of the corresponding oxalates was formed. The decomposition of the precipitate and the solid-state reaction were performed by heating at 900 °C for 5 h. The product was pressed into pellets at 4 kbar, and reheated to 900 °C for sintering.

#### 2. X-Ray Analysis

X-ray powder diffractograms (System D 500 SIE-MENS) revealed three individual crystallographic phases. Within a range of 10° to 80° (2 $\theta$ ), 17 lines could be identified to correspond to a layer-type perovskite-like phase, related to the K2NiF4 structure (a=3.79 Å and c=13.21 Å) [16]. The second phase is most probably a cubic one, whose presence depends on the Ba concentration, as the line intensity decreases for smaller x(Ba). The amount of the third phase (volume fraction > 30% from the x-ray intensities) seems to be independent of the starting composition, and shows thermal stability up to 1,000 °C. For higher temperatures, this phase disappears progressively, giving rise to the formation of an oxygen-deficient perovskite (La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14</sub>) as described by Michel and Raveau [16].

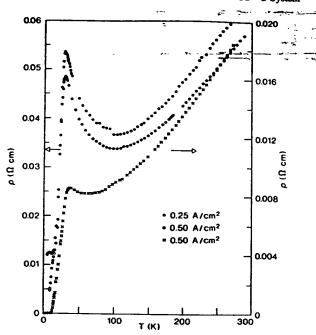


Fig. 1. Temperature dependence of resistivity in  $Ba_xLa_{5-x}Cu_5O_{5(3-p)}$  for samples with x(Ba)=1 (upper curves, left scale) and x(Ba)=0.75 (lower curve, right scale). The first two cases also show the influence of current density

#### 3. Conductivity Measurements

The dc conductivity was measured by the four-point method. Rectangular-shaped samples, cut from the sintered pellets, were provided with gold electrodes and contacted by In wires. Our measurements between 300 and 4.2 K were performed in a continuous-flow cryostat (Leybold-Hereaus) incorporated in a computer-controlled (IBM-PC) fully-automatic system for temperature variation, data acquisition and processing.

For samples with  $x(Ba) \le 1.0$ , the conductivity measurements, involving typical current densities of 0.5 A/cm<sup>2</sup>, generally exhibit a high-temperature metallic behaviour with an increase in resistivity at low temperatures (Fig. 1). At still lower temperatures, a sharp drop in resistivity (>90%) occurs, which for higher currents becomes partially suppressed (Fig. 1: upper curves, left scale). This characteristic drop has been studied as a function of annealing conditions, i.e., temperature and O<sub>2</sub> partial pressure (Fig. 2). For samples annealed in air, the transition from itinerant to localized behaviour, as indicated by the minimum in resistivity in the 80 K range, is not very pronounced. Annealing in a slightly reducing atmosphere, however, leads to an increase in resistivity and a more pronounced localization effect. At the same time, the onset of the resistivity drop is shifted

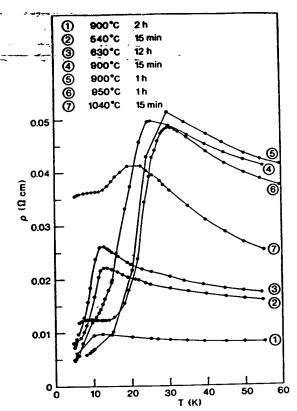


Fig. 2. Low-temperature resistivity of samples with x(Ba) = 1.0, annealed at  $O_2$  partial pressure of 0.2 bar (curve ①) and  $0.2 \times 10^{-4}$  bar (curves ② to ⑦)

towards the 30 K region. Curves @ and ⑤, recorded for samples treated at 900 °C, show the occurrence of a shoulder at still lower temperature, more pronounced in curve 6. At annealing temperatures of 1,040 °C, the highly conducting phase has almost vanished. As mentioned in the Introduction, the mixed-valent state of copper is of importance for electron-phonon coupling. Therefore, the concentration of electrons was varied by the Ba/La ratio. A typical curve for a sample with a lower Ba concentration of 0.75 is shown in Fig. 1 (right scale). Its resistivity decreases by at least three orders of magnitude, giving evidence for the bulk being superconducting below 13 K with an onset around 35 K, as shown in Fig. 3, on an expanded temperature scale. The latter figure also shows the influence of the current density, typical for granular compounds.

#### III. Discussion

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The resistivity behaviour of our samples, Fig. 1. is qualitatively very similar to the one reported in the Li-Ti-O system, and in superconducting

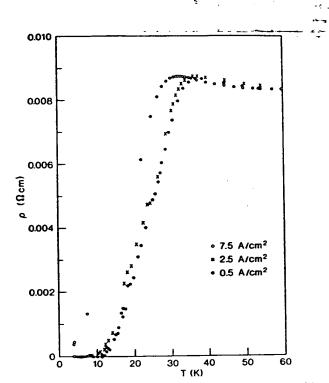


Fig. 3. Low-temperature resistivity of a sample with x(Ba) = 0.75, recorded for different current densities

BaPb<sub>1-x</sub>Bi<sub>x</sub>O<sub>3</sub> polycrystalline thin films [5, 18]. Upon cooling from room temperature, the latter exhibit a nearly linear metallic decrease of  $\rho(T)$ , then a logarithmic type of increase, before undergoing the transition to superconductivity. One could, of course, speculate that in our samples a metal-to-metal structural phase transition occurs in one of the phases. The shift in the drop in  $\rho(T)$  with increasing current density (Fig. 3), however, would be hard to explain with such an assumption, while it supports our interpretation that we observe the onset of superconductivity of percolative nature, as discussed below. In BaPb<sub>1-x</sub>Bi<sub>x</sub>O<sub>3</sub>, the onset of superconductivity has been taken at the resistivity peak [18]. This assumption appears to be valid in percolative systems, i.e., in the thin films [18] consisting of polycrystals with grain boundaries, or when different crystalline phases with interpenetrating grains are present, as found in the Li-Ti-O [5] or in our Ba-La-Cu-O system. The onset can also be due to fluctuations in the superconducting wave functions. We assume one of the Ba-La-Cu-O phases exhibits this behaviour. Therefore, under the above premises, the peak in  $\rho(T)$ at 35 K, observed for an x(Ba) = 0.75 (Fig. 1), has to be identified as the start to superconductive cooperative phenomena in the isolated grains. It should be noted that in granular Al, Cooper pairs in coupled grains have been shown to exist already at a point where  $\rho(T)$  upon cooling has decreased by only 20% of its highest value. This has been proven qualitatively [19] and more recently also quantitatively [20] by the negative frequency shift occurring in a microwave cavity. In 100 Å films, a shoulder in the frequency shift owing to 2D fluctuations was observed above the  $T_c$  of the grains. In our Ba - La - Cu - O system, a series of layer-like phases with considerable variety in compositions are known to exist [16, 21], and therefore 2D correlations can be present.

The granularity of our system can be justified from the structural information, and more quantitatively from the normal conductivity behaviour. From the former, we know that more than one phase is present and the question arises how large are the grains. This can be inferred from the logarithmic fingerprint in resistivity. Such logarithmic increases are usually associated with beginning of localization. A most recent example is the Anderson transition in granular Sn films [22]. Common for the granular Sn and our samples is also the resistivity at 300 K, lying in the range of 0.06 to 0.02  $\Omega$ cm, which is near the microscopic critical resistivity of  $\rho_c = 10 L_0 \hbar/e^2$ for localization. From the latter formula, an interatomic distance  $L_0$  in the range of 100 Å is computed, thus a size of superconducting grains of this order of magnitude must be present. Upon cooling below  $T_c$ , Josephson junctions between the grains phaselock progressively [23] and the bulk resistivity gradually drops to zero by three orders of magnitude, for sample 2 (Fig. 1). At larger current densities, the weaker Josephson junctions switch to normal resistivity, resulting in a temperature shift of the drop, as shown in Fig. 3. The plateau in resistivity occurring below the 80% drop (Fig. 1) for the higher current density of 0.5 A/cm<sup>2</sup>, and Fig. 2 curve (6) may be ascribed to switching of junctions to the normal state.

The way the samples have been prepared seems to be of crucial importance: Michel et al. [21] obtained a single-phase perovskite by mixing the oxides of La and Cu and BaCO<sub>3</sub> in an appropriate ratio and subsequent annealing at 1,000 °C in air. We also applied this annealing condition to one of our samples, obtained by the decomposition of the corresponding oxalates, and found no superconductivity. Thus, the preparation from the oxalates and annealing below 950 °C are necessary to obtain a non-perovskite-type phase with a limited temperature range of stability exhibiting this new behaviour. The formation of this phase at comparatively low temperatures is favoured by the intimate mixture of the compo-

nents and the high reactivity of the oxalates owing to the evolution of large amounts of H<sub>2</sub>O-and CO<sub>2</sub> during decomposition.

#### IV. Conclusion

In the concentration range investigated, compounds of the Ba-La-Cu-O system are metallic at high temperatures, and exhibit a tendency towards localization upon cooling. Samples annealed near 900 °C under reducing conditions show features associated with an onset of granular superconductivity near 30 K. The system consists of three phases, one of them having a metallic perovskite-type layer-like structure. The characterization of the new, apparently superconducting, phase is in progress. An identification of that phase may allow growing of single crystals for studying the Meissner effect, and collecting specific-heat data to prove the presence of high  $T_c$  bulk superconductivity.

The authors would like to thank H.E. Weibel for his help in getting familiar with the conductivity measurement system, E. Courtens and H. Thomas for discussions and a critical reading of the manuscript.

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#### J.G. Bednorz and K.A. Müller: Ba-La-Cu-O System

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#### Note Added in Proof

Chemical analysis of the bulk composition of our samples revealed a deviation from the ideal La/Ba ratios of 4 and 5.66. The actual ratios are 16 and 18, respectively. This is in agreement with an identification of the third phase as CuO.



#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J. Bednorz et al.

Date: March 6, 1997

Serial No.: 08/303,561

Group Art Unit: 1105

Filed: September 9, 1994

Examiner: D. McGinty

For:

NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH

TRANSITION TEMPERATURE, AND METHODS FOR THEIR

**USE AND PREPARATION** 

The Commissioner of Patents and Trademarks Washington, D.C.

### SUBSTITUTE AMENDMENT

Sir:

In response to the Office Letter dated January 8, 1997, please consider the following:

## IN THE CLAIMS

Add claims 114-128.

114. (Added) A method including the steps of forming copper oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26°K;

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said

• •

superconducting state;

said copper oxide includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element.

115. (Added) A method comprising the steps of:

forming a composition including copper, oxygen and an element selected from the group consisting of at least one Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, where said composition is a mixed copper oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K;

maintaining said composition in said superconducting state at a temperature greater than 26°K; and

passing an electrical current through said composition while said composition is in said superconducting state.

116. (Added) A method including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of 26°K;

maintaining said composition at a temperature in excess of 26°K at which temperature

said composition exhibits said superconductive state;

passing an electrical current through said composition while said composition is in said superconductive state; and

said composition including a copper oxide and at least one element selected from the group consisting of Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element.

117. (Added) A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:

- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element;
- (b) maintaining the superconductor element at a temperature above  $26^{\circ}\text{K}$  and below the superconductor transition temperature  $T_c$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

118. (Added) A superconductive method for conducting an electric current essentially without resistive losses, comprising:

- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the copper-oxide compound including at least one element selected from the group consisting of a group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{c}$  the transition-onset temperature  $T_c$  being greater than 26°K;
- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{\rho=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 119. (Added) A method including the steps of forming a transition metal oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26°K;

maintaining the temperature of said material at a temperature less than said critical

temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said superconducting state;

said transitional metal oxide includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element.

120. (Added) A method comprising the steps of:

forming a composition including a transition metal, oxygen and an element selected from the group consisting of at least one Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, where said composition is a mixed transitional metal oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K;

maintaining said composition in said superconducting state at a temperature greater than 26°K; and

passing an electrical current through said composition while said composition is in said superconducting state.

121. (Added) A method including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of

26°K;

maintaining said composition at a temperature in excess of 26°K at which temperature said composition exhibits said superconductive state;

passing an electrical current through said composition while said composition is in said superconductive state; and

said composition including a transitional metal oxide and at least one element selected from the group consisting of Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element.

122. (Added) A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:

- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a transition metal oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductive transition temperature  $T_c$  of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element;
- (b) maintaining the superconductor element at a temperature above  $26^{\circ}\text{K}$  and below the superconductor transition temperature  $T_c$  of the superconductive composition; and

(c) causing an electric current to flow in the superconductor element.

123. (Added) A superconductive method for conducting an electric current essentially without resistive losses, comprising:

- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a transition metal-oxide compound having a layer-type perovskite-like crystal structure, the transition metal-oxide compound including at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{\rho=0}$ , the transition-onset temperature  $T_c$  being greater than  $26^{\circ}K$ ;
- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{\rho=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 124. (Added) A method including the steps of forming copper oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26°K;

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said superconducting state;

said copper oxide includes at least one element selected from the group consisting of a Group II A element, at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element.

125. (Added) A method comprising the steps of:

forming a composition including copper, oxygen and any element selected from the group consisting of at least one Group II A element and at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element, where said composition is a mixed copper oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K;

maintaining said composition in said superconducting state at a temperature greater than 26°K; and

passing an electrical current through said composition while said composition is in said superconducting state.

126. (Added) A method including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of

26°K;

maintaining said composition at a temperature in excess of 26°K at which temperature said composition exhibits said superconductive state;

passing an electrical current through said composition while said composition is in said superconductive state; and

said composition including a copper oxide and at least one element selected from the group consisting of Group II A element, at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element.

127. (Added) A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:

- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductive transition temperature  $T_c$  of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element, at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element;
- (b) maintaining the superconductor element at a temperature above

 $26^{\circ}\text{K}$  and below the superconductor transition temperature  $T_{\text{c}}$  of the superconductive composition; and

(c) causing an electric current to flow in the superconductor element.

128. (Added) A superconductive method for conducting an electric current essentially without resistive losses, comprising:

(a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the copper-oxide compound including at least one element selected from the group consisting of a group II A element, at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature T<sub>c</sub> and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature

 $T_{\rho=0}$ , the transition-onset temperature  $T_c$  being greater than 26°K;

- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{\rho=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

#### REMARKS

Claims 24-26, 86-90 and 96 to 128 are in the application.

Claims 113-128 are added by this amendment.

Herein, EA will refer to the Examiner's Action's dated April 15, 1996.

In regard to applicant's claim of priority, in EA paragraphs 3.b.i, 3.b.ii, 3.b.iv and 3.v the Examiner states "the certified priority document may provide basis for compositions of the formula  $RE_2TM.O_4$ ". It is noted that the Abstract of the priority document refers to "the general formula  $RE_{2.x}AE_xTMO_{4.y}$ , wherein RE is a rare earth, AE is a member of the group of alkaline earths or a combination of at least two members of that group, and TM is a transition metal, and wherein x < 0.3 and  $0.1 \le y \le 0.5$ ." This formula permits no alkaline earth and a varying amount of alkaline earth and rare earths and a varying amount of oxygen. At column 3, lines 20 and 35, there is recited "the Ba-La-Cu-O system" and at line 41 "La<sub>2.x</sub>Ba<sub>x</sub>DuO<sub>4.y</sub> x < 1 and  $y \le 0$  and at line 44 teaches La<sub>1.x</sub>Ba<sub>x</sub>CuO<sub>3.y</sub>."

The Examiner at page 2 of PA at paragraph 3.b.i states that the priority document does not provide support for "the limitations a composition including a transition metal, a rare earth or rare earth-like elements, an alkaline earth element, an oxygen as found in claim 86". It is noted that in the priority document at column 2, lines 13-19 it is stated that "it is a characteristic of the present invention that in the compounds in question that the RE portion is partially substituted by one member of the alkaline earth group of metals, or by a combination of the members of this alkaline

earth group and that the oxygen content is at a deficit." It is further noted that at column 2, lines 20-23 it states that "for example, one such compound that meets the description given by this lanthanum copper oxide La<sub>2</sub>CuO<sub>4</sub> in which the tantalum which belongs to the IIIB group of the elements is in part substituted by one member of the neighboring IIIA group of elements." In the sentence bridging pages 2 and 3 of EA, the Examiner states that "the certified priority document may provide a basis for formula RE<sub>2</sub>TM.O<sub>4</sub> at P.2, para. 4, but the claimed composition is deemed to be much broader than that formula." It is clear from the quoted sections of the priority document that the priority document clearly supports a much broader composition than the Examiner is stating that the priority document supports, and that the priority document, in fact, does support applicants' claim 86.

At page 3, paragraph ii of EA, the Examiner states there is no support for "the limitation 'non-stoichiometric amount of oxygen', as found in claim 86". Applicants submit that the use of the term oxygen deficit is noted by the Examiner at P.2, para. 4 of the priority document and the varying amount of oxygen given in both formulas is sufficient and adequate support for the limitation 'non-stoichiometric' amount of oxygen." In regards to paragraph iii of page 3 of EA, the Examiner states there is no support for "transition metal oxides" as found in claim 24. Claim 24 recites transition metal oxide which is explicitly recited in the priority document, as stated above. Claim 88 is directed to the superconducting material having a transition temperature in excess of 26°K. Claim 89 depends from claim 88 and recites that "said composition is comprised of a metal oxide." The priority document supports superconducting material containing or comprising a metal oxide. Claim 90 depends from claim 88 and recites "where said composition is comprised of a transition metal oxide, a copper oxide is a transition metal oxide." The published patent application corresponding to the priority document (EPO 0 274 343 A1, 7-27-88) at column 3, line 6 recites Ti as a transition

metal. It is noted that in claim 1 of the EPO published patent application corresponding to the priority document, claim 1 recites the structure RE<sub>2-x</sub>AE<sub>x</sub>TM.O<sub>4-y</sub> wherein TM is a transition metal. Claim 2 therein recites copper as the transition metal. Claim 3 therein recites nickel as the transition metal. Claim 8 therein recites chromium as the transition metal. Consequently, a broader class of transition metals other than copper is supported by the priority document.

At paragraph iv, on page 3 of PA, the Examiner states that "the limitation of 'copper-oxide compounds', as recited in claim 96" is not supported by the priority document with regards to which the Examiner states "the certified priority document may provide basis for compositions of the formula RE<sub>2</sub>TM.O<sub>4</sub>." As noted above, the general formula recited by the Examiner is incorrectly stated and should be stated wherein the quantity of oxygen, of the rare earth element and of an alkaline element is variable. Consequently, the term "a copper-oxide compound" is adequately supported by the priority document.

In paragraph v on page 3 of EA, the Examiner states that "the limitation to the effect that the 'copper-oxide compound' includes (including) at least one rare earth, rare-earth-like element and at least one alkaline-earth element 'as recited in claim 97... at claim 103...' is not supported by the priority document." The Examiner further states "the certified priority document may provide basis for compositions of the formula RE<sub>2</sub>TM.O<sub>4</sub>". Applicants as stated above respectfully submit the priority document refers throughout and, in particular, in the Abstract to "the general formula RE<sub>2-x</sub>AE<sub>x</sub>EM.O<sub>4-y</sub> as stated above which includes a copper-oxide as stated above. The Examiner further states "but basis is not seen for the more general limitation of 'a copper-oxide compound' with a rare-earth (like) element and in alkaline earth element." It is noted that in the priority document, claim 2 refers to lanthanum as the

rare earth; claim 3 refers to cerium as the rare earth; claim 5 refers to barium as a partial substitute for the rare earth; claim 6 refers to calcium as a partial substitute for the rare earth; claim 7 refers to strontium as a partial substitute for the rare earth and claim 9 refers to neodymium as the rare earth. Clearly, priority document teaches barium, calcium, strontium. Consequently, the priority document supports the term rare earth-like since there are other elements other than those commonly referred to as the rare earth which are elements 57-71 which satisfy the teaching of the priority document and of the present application. The Abstract of the priority document refers to "AE as a member of the alkaline earth or a combination of at least two members of that group". Consequently, the priority document clearly supports an alkaline earth element.

At paragraph vi of page 4 of EA, the Examiner asserts that the priority document does not support "a non-stoichiometric atomic proportion" as found in claims 101, 102, 107 and 108. The exemplary general formula recited above which is recited in the priority document clearly shows the oxygen has a variable content and, consequently, is not in stoichiometric proportion. Consequently, the priority document clearly supports the term "non-stoichiometric atomic proportion".

At paragraph vii of page 4 of EA, the Examiner states that the priority document does not support "the limitation as to 'the effectively-zero-bulk resistivity intercept temperature T<sub>0</sub>', as found in claim 103." Applicants responded to this same comment at page 6 of their response dated September 26, 1995 at pages 4-6 thereof. The Examiner has not commented upon applicant's remarks and has merely repeated what was said in the Examiner's prior Office Action. Applicant's assume that the Examiner agrees with applicant's statements in their prior response in that the concept of the intercept temperature is well known in the prior art and can be included in claim

103.

In view of the remarks herein, the Examiner is respectfully requested to withdraw the objection to applicant's claim for priority under 35 USC 119 based on applicant's priority document.

In paragraph 4 of EA at page 4 thereof, the specification has been objected to under 35 USC 112, first paragraph, as failing to provide an enabling disclosure commensurate with the scope of the claims. Paragraphs 4.A, 4.B and 4.C on pages 4-5 of the PA are identical to the Examiner's comments in the previous Office Action.

Paragraph 4-C on page 5 of EA rejects claims 24-26, 86-90 and 96-108 under 35 USC 112, first paragraph, for the same reasons set forth in the objection to the specification which is the same as in the Examiner's prior Office Action.

In paragraph 4-D at page 5 of EA, the Examiner states that he has considered applicant's arguments in response to the Examiner's prior Office Action stating that they "have been fully considered but they are not deemed to be persuasive." At paragraph 4.D.i at page 5 of EA, the Examiner states "the additional case law and arguments by the applicants have been newly noted. For the reasons that follow, however, the record as a whole is deemed to support the initial determination that the originally filed disclosure would not have enabled one skilled in the art to make and use the invention to the scope that it presently claimed." The Examiner does not specifically respond to the specific passage cited from the case law, nor rebut their applicability in the way applicant's have applied them. Consequently, the Examiner's silence is viewed as agreement with applicant's argument.

At paragraph d.ii on page 6 of EA, the Examiner states that "the applicant's quote several passages from their specification at pp. 13-15 of their September 29, 1995 amendment, the issue is the scope of enablement, not support". The Examiner further states "the issue here is the scope to which that disclosure would have taught one skilled in the art how to make and use the composition which shows the onset of superconductivity above 26°K." Applicant's respectfully disagree since recitation of examples is part of the support for the scope of enablement. It addition to the examples recited at page 13-15 of applicant's specification, applicants' comments in their September 29, 1995 amendment, at pages 15-25 clearly show that applicant's "disclosure would have taught one skilled in the art how to make and use the composition which shows the onset of superconductivity above 26°K."

At paragraph d.iii on page 6 of EA, the Examiner states "construed in light of that issue, the invention is not deemed to have been fully enabled by the disclosure to the extent fully claimed." Applicant's respectfully disagree and note that the Examiner has not specifically rebutted applicant's arguments on page 15-25 of applicant's September 29, 1995 amendment. All that the Examiner has said is that "the invention is not deemed to have been fully enabled by the disclosure to the extent fully claimed."

At paragraph d.iii.1 on page 6 of EA, the Examiner states in regard to applicant's argument in their September 29, 1995 amendment that applicant's states their disclosure "lists several species such as La<sub>2-x</sub>Ba<sub>x</sub>CuO<sub>4-y</sub> which they indicate are found in the present disclosure." Applicant's disclosure supports a substantially broader scope than this species. In particular, the Examiner is directed to applicant's Summary of Invention on page 6-9 of applicant's application. There is no requirement that applicants list every possible species that could possibly come within the scope of

applicant's claims. Applicant's broadly teach transition metal oxides which can contain rare earth and alkaline earth elements.

At paragraph D.iii.1.a, at page 6 of EA, the Examiner states "notwithstanding that argument it still does not follow that the invention is fully enabled for the scope presently claimed." Applicant's respectfully disagree for the reasons provided by applicants on page 15-25 of their September 29, 1995 amendment.

At paragraph d.iii.1.b at page 6 of EA, the Examiner refers to the paragraph bridging pages 3 and 4 of applicant's specification. The Examiner states "the present specification actually shows that known forms of 'transition metal oxide' and 'a copper-oxide compound' do not show the onset of superconductivity above 26°. The Examiner then states that "applicants state that the prior art includes a Li-Ti-O system with superconducting onsets as high as 13.7° K." Applicants do not see the relevance of the Examiner's statements. Such a composition would not be included within the claims since applicant's claim covers only compositions having superconductivity above 26°K. Applicant's acknowledge that Ti is a transition metal. The Examiner notes "that disclosure also refers to 'a second non-conducting CuO phase at p. 14, line 18." Applicant's do not understand the significance of this quoted passage is to the Examiner's argument.

At paragraph d.iii.1.c at page 7 of EA, the Examiner states "accordingly, the present disclosure is not deemed to have been fully enabling with respect to the 'transition metal oxide' of claim 24, the 'composition' of claim 88 or the 'copper-oxide compound' of claim 98." Again, applicant's note applicant's arguments on page 15-25 of their September 29, 1995 amendment. Applicant's, at page 5, line 3 of their specification, refer to transition metal oxides and the sentence bridging pages 5 and 6

to superconducting composition at a T<sub>c</sub> greater than 26°K.

At paragraph d.iii.2 of page 7 of EA, the Examiner states that "the examples of p. 18, lines 1-20, of the present specification further substantiate the finding that the invention is not fully enabled for the scope presently claimed." Applicant's respectfully disagree.

At paragraph d.iii.2.a at page 7 of EA, the Examiner refers to an example in the first paragraph of page 18 of their specification which says at line 10 "and there is no superconductivity." The Examiner appears to be using this paragraph to support the Examiner's assertion that applicant's claims are not enabled by their disclosure. Quite to the contrary, this paragraph supports applicant's assertion that their claims are enabled. Applicant's are providing a broad teaching of how these compositions can be fabricated, by providing a teaching which has not resulted in superconductivity, applicant's are providing a teaching of methods which do lead to examples showing superconductivity. Even if the claims encompass some inoperative examples, this does not render the claims unenabled. Moreover, the claims specifically refer to compounds which are superconducting. Consequently, a sample which is not superconducting is not within the scope of the claim. Applicant's submit that the Examiner is citing fragments of statements from their specification out of context resulting in a misunderstanding of applicant's teaching.

At paragraph d.iii.2.b, at page 7 of EA, the Examiner refers to applicant's example which appears to be in the third paragraph of page 18 of applicant's specification which at line 20 recites  $T_c$ =26°K. The Examiner then says that applicant's claims require  $T_c$  to be greater than 26°K in what appears to be an attempt to show that applicant's claims are not enabled. Applicants do not believe the

recitation of 26°K in the specification and > 26°K in the claims has any significance to this argument. Applicant's can amend their claims to say ≥ 26° if that's what the Examiner would prefer. Clearly, the temperature consistent with applicant's claims can be infinitesimally close to 26°K.

At paragraph d.ii.2.c at page 7 of EA, the Examiner states "consequently, the present disclosure is not deemed to adequately enable the full scope of the present claims." The Examiner further states "independent claims 86 and 103 may require the presence of rare earth, alkaline earth, and transition metals, but the aforementioned examples show that superconductivity is still very unpredictable." Applicant's respectfully disagree that the aforementioned examples show that superconductivity is still very unpredictable. The Examiner has taken applicant's examples out of context. These examples are provided as part of the teaching on how to fabricate the claimed invention.

The Examiner further states "those claims cannot be deemed to be fully enabled." Applicants respectfully disagree. It is also noted again that the Examiner has not addressed applicants arguments on page 15-25 of applicants September 29, 1995 amendment.

At paragraph d.iv on page 7 of EA, the Examiner refers to 3 affidavits submitted by applicants. Applicants acknowledge that the 3 affiants are employees of the assignee of the present application. At paragraph d.IV.1 at page 8 of EA the Examiner states "those affidavits do not set forth particular facts to support the conclusions that all superconductors based on applicants' work behave in the same way and that one of skill in the art can make those superconductors without undue experimentation. Conclusionary statements in an affidavit or specification do not

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provide the facts or evidence needed for patentability." The referred to affidavits are dated after August 19, 1995 a period of more than 8 years after the present application was filed. Those affidavits refer to developments in the field after the publication of applicants which was cited on page 6 of applicants specification. The statements in the affidavits are not conclusionary but are statements of fact. By the Examiners statement that these are conclusionary the Examiner appears to be placing himself up as an expert in the field of superconductivity. Applicants respectfully request that the Examiner submit an affidavit in the present application rebutting the position taken by applicants 3 affiants.

At paragraph d.iv.2, at page 8 of PA, the Examiner states "those affidavits do not overcome the non-enablement rejection. The present specification discloses on its face that only certain oxides compositions of rare earth, alkaline earth and transition metals made according to the certain steps will superconduct at greater than 26°K." Applicants respectfully disagree with this statement. Applicants' specifications discloses substantially more as applicants have indicated above and as applicants have indicated in their amendment of September 29, 1995. Applicants work clearly started the field of high-temperature superconductivity. Consequently, applicants teaching has enabled this entire field. The Examiners statements to the contrary have no basis in fact.

At paragraph d.iv.3 of page 8 of PA, the Examiner states "those affidavits are not deemed to shed light on the state of the art and enablement at the time the invention was made." Applicant's respectfully disagree. The Examiner has not shown any reason contrary to applicants assertion that the superconducting materials can be made by the methods disclosed by applicant's and as stated by applicant's 3 affiants. Applicant's have objectively enabled their application and their claims. Applicant's

have pointed to copious locations in their specification which do provide support for applicant's claims.

At paragraph d.iv.4 at page 8 of EA, the Examiner states that "it is fully understood that the applicant's are the pioneers in high temperature metal-oxide superconductivity. The finding remains, nonetheless, that the disclosure is not fully enabling for the scope of the present claims." Applicant's respectfully disagree. The Examiner has provided no substantial evidence to support this assertion. It is respectfully requested that the Examiner support their assertion with factual evidence and not unsupported statements.

In view of the remarks herein, the Examiner is respectfully requested to withdraw the objection to the specification under 35 USC 112, first paragraph, and the rejection of claims 24-26, 86-90 and 96-108 under 35 USC 112, first paragraph.

Claims 86-87 and 96-108 have been rejected under 35 USC 112, second paragraph as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant's regard as their invention. Applicant's note that the Examiner has not responded to applicant's comments which supports applicants position that a person of skill in the art would understand the terms "layer-type" and "perovskite-like" and has just repeated their rejection of the prior Office Action.

Applicant's respectfully request the Examiner to comment on applicant's prior remarks.

In view of the remarks herein, the Examiner is respectfully requested to withdraw the rejection of claims 86-87 and 96-108 under 35 USC 112, second paragraph.

Claims 24-26, 86-90 and 96-108 have been rejected under 35 USC 102(a) as being anticipated by Asahi Shinbum Int'l. Satellite Edition (London) November 11, 1986. The Examiner incorrectly gives a date of November 11, 1986 for this article. It is November 28, 1986.

Paragraph 6.a and 6.b of page 10 of EA are essentially the same as in the Examiner's prior action. Applicant's have responded to these paragraphs in their prior response.

In paragraph 6-C at page 10 of EA, the Examiner notes applicant's prior responses and states that they "have been fully considered but they are not deemed to be persuasive." It is noted that in the declaration of co-inventors J.G. Bednorz and K.A. Mueller dated March 21, 1988, mailed into the patent office on June 22, 1988 at paragraph 3 states "On approximately October 16, 1986, we gave Praveen Chaudhari... six samples of the high temperature superconductive ceramic oxide materials that we had described in our aforementioned Z Physik B. publication. Praveen Chaudhari brought these samples back to the U.S. when he returned after visiting with us on or about October 16, 1986." This is evidence that these samples are brought into the United States on or about October 16, 1986. When these samples came into the United States, since they were inherently superconductive as claimed, the invention was essentially reduced to practice in the United States on that date. It is further noted that the Declaration of Alexis P. Malozenoff signed March 30, 1988 states at paragraph 3, "On or about November 15, 1986, Richard Greene and I travelled to Baltimore for a magnetics conference. During our travel to Baltimore, we discussed Greene's ongoing experiments in high T<sub>c</sub> superconducting samples which he said had been received from Bednorz and Mueller." This is clear evidence that by November 15, 1986, superconducting samples fabricated by applicant's were being

measured in the United States. These samples were inherently superconducting and, consequently, established the reduction to practice in the United States as of that date. The Declaration of Cheng-Chung John Chi dated March 29, 1988 states at paragraph 2, "At a time prior to approximately the middle of November, 1986, Chang C. Tsuei told me a measurement he made on T<sub>c</sub> superconducting material which he said were received from Georg Bednorz and K.A. Mueller, two physicists working for IBM Corporation in Zurich, Switzerland... Chang Tsuei said that he had measured resistivity versus temperature of these samples." This is again further evidence that the Mueller Bednorz superconducting samples were in the United States prior to the middle of November 1986."

At page 11 of EA in the paragraph labelled i, the Examiner states "the applicants argue that Sung II Park affidavit of March 30, 1988 states at para. 4 that measurements were taken of a superconductive sample on or before November 9, 1986, to the best of affiants recollection, or no later than November 15, 1986. The document evidence is not deemed to support that argument, however." In the paragraph marked (1) on page 11 of PA, the Examiner states "plots of those measurements are missing. See the Cheng C. Tseui affidavit of March 30, 1988, para. 6." This statement comes directly out of Cheng Tseui's declaration. Notwithstanding, Cheng Tseui's declaration says the measurements were made, that the plots that were taken were missing. The last sentence of this paragraph states "I believe that they may have been inadvertently thrown away when the laboratory was subsequently extensively cleaned." The Examiner further states "a hand-drawn diagram with the indication of a vacuum pumped down on November 9, 1988 also is not deemed to show that the measurements were taken." The Examiner is referring to paragraph 5 of the Cheng Tseui declaration and exhibit C which contains the handdrawn figure.

At paragraph (2) of page 11 of EA, the Examiner points to cablegrams sent by Dr. Greene to applicants in Zurich which are attached as exhibit B to his declaration. The Examiner states "Dr. Greene reports that no indication of superconductivity has been seen in his specific heat measurements for temperature 4-35°K." The Examiner fails to note that in the same cablegram dated November 11, 1986, Dr. Greene states "this is not really too surprising given the very broad transition to have found in resistivity and susceptibility." The Examiner acknowledges that "exhibit C has pages dated December 1, 1986 on in exhibit D, which actually has plots and resistance versus temperature dated as early as December 3, 1986." The Examiner is conceding that high T<sub>c</sub> superconductivity was measured on the samples which the very same set of cablegrams and affidavit say were in the United States in the middle of November 1986. Consequently, by the Examiner's own admission, samples which were in the United States were clearly shown to be superconducting as of December 3, 1986. Consequently, the samples that were in the United States as of November 9 were inherently superconducting. It is clear from the same declarations that applicant's were communicating with Dr. Greene. It is noted that Dr. Greene's cablegram dated November 25, 1986 to applicants states he will resume work on the new superconductor and that not much will happen because of the Thanksgiving holiday until the following week. There are cablegrams dated November 26, December 1, December 2, 1986 related to high T<sub>c</sub> superconductivity. Dr. Greene's exhibit C has notebook pages dated December 1, 1986 to December 5, 1986. The December 5, 1986 shows  $T_c$  of 26°K and 30°K. Exhibit D show a plot of R vs. T dated December 8, 1986. Clear reduction to practice is shown and clear diligence is shown from prior to the date of the Asahi Shinbum article. This was clearly done in close correspondence with the applicants. Thus, the facts clearly show applicant's can swear behind the Asahi Shinbum reference.

At paragraph ii on page 11 of EA, the Examiner states "the applicant's assert that the Asahi Shinbum article reports a third parties confirmation of their original discovery. That assertion appears to be correct, but the article is still deemed to be prior art under 35 USC 102(a). At page 12 under paragraph 2, subparagraphs A, B and C, the Examiner made comments in regards to four cases applicant's have cited in support of their position that the Asahi Shinbum article should not be prior art because to hold it as prior art would not afford applicant's the benefit the one year grace period provided them under 35 USC 102(b).

At paragraph 3 on page 13 of EA, applicant's respectfully disagree that the earliest date with which applicant's can show for their invention in this country is December 1986. Numerous affidavits which applicant's have submitted clearly show that applicant's have, in early November 1986, the superconducting compounds which the Examiner admits in applicant's data of December 3, 1986 shown the measurements of critical temperatures. Consequently, the Examiner's statement acknowledges the Examiner's apparent agreement in the fact that the materials were in this country in the middle of November 1986. Applicant's respectfully disagree with the Examiner's statement, "notwithstanding the possible uniqueness of the present facts, however, the Asahi Shinbum article is still deemed to be prior art under 35 USC 102(a), which the applicant's have not been able to overcome with a showing of early date in this country or showing of their direction and control over the work done by the third party." Applicant's note that the Asahi Shinbum article provides no enablement but merely is an assertion of a result achieved which points to applicant's own work which was reported in the article applicant's cite in their application at page 6. Consequently, any description in the Asahi Shinbum article is applicant's own work. If one would follow the rationale of the Examiner, if an applicant publishes an article and

some other third party reports that same result prior to applicant's filing of a patent application, (which is subsequently filed within one year of applicant's own publication) the reporting of applicant's work by the third party would be prior art against applicant's application. Such a result would deny applicant's the one year grace period provided under 35 USC 102(b).

In view of the changes to the claims and the remarks herein, the Examiner is respectfully requested to reconsider the above-identified application. If the Examiner wishes to discuss the application further, or if additional information would be required, the undersigned will cooperate fully to assist in the prosecution of this application.

Please charge any fee necessary to enter this paper to deposit account 09-0468.

If the above-identified Examiner's Action is a final Action, and if the above-identified application will be abandoned without further action by applicants, applicants file a Notice of Appeal to the Board of Appeals and Interferences appealing the final rejection of the claims in the above-identified Examiner's Action. Please charge deposit account 09-0468 any fee necessary to enter such Notice of Appeal.

Respectfully submitted,

Daniel P. Morris Reg. No. 32,053

IBM Corporation Intellectual Property Law Dept. P.O. Box 218 Yorktown Heights, N.Y. 10598 (914) 945-3216 Enteri amendment was sent.

### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants:

J. Bednorz et al.

Date: February 10, 1997

Serial No.: 08/303,561

Group Art Unit: 1105

Filed: September 9, 1994

Examiner: D. McGinty

For:

NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH

TRANSITION TEMPERATURE, AND METHODS FOR THEIR

**USE AND PREPARATION** 

The Commissioner of Patents and Trademarks Washington, D.C.

#### **AMENDMENT**

Sir:

In response to the Office Letter dated January 8, 1997, please consider the following:

#### IN THE CLAIMS

Add claims 114-122.

114. (Added) A method including the steps of forming collaboration which exhibits a superconducting state at a critica 26°K;

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maintaining the temperature of said material at a temperature temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said

Mendix C

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# THEORY OF SUPERCONDUCTIVITY

Вч

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New York, 1952

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#### **Fundamental Facts**

(a) Superconductivity was discovered in 1911 by Kamerlingh-Onnes. He was the first to liquefy helium and so to produce temperatures below 10° K. With this new technique he was able to observe the continued decrease of the electrical resistance of metals with decreasing temperature. With mercury, in contrast to other metals, he was astonished to find that the resistance completely vanished, almost discontinuously, at about 4.2° K

(Fig. 1-1). Today superconductivity is known in 18 other metals (see Table 1-1) whereas in others, e. g., gold and bismuth, the conductivity remains normal far below even 1°K. Many alloys and compounds can also become superconducting, in particular the frequently used niobium nitride which has a transition temperature as high as 20°K. However, among these latter substances hysteresis phenomena mentioned in the "Introduction" are so much more strongly evident that in testing the present theory we prefer to employ only the "good" superconductors, i. e., the pure elements.

In the ideal case the resistance vanishes completely and discontinuously at a transition temperature  $T_s$ . Actually the resistance-temperature curve does fall more sharply the more the specimen is like a single crystal and the smaller the measuring current used. Because the drop always occurs in a measurable temperature range, the experimental definition of the transition temperature is to some extent arbitrary. The temperature at

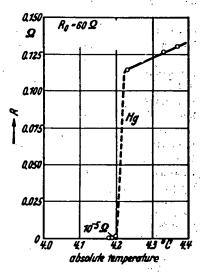


Fig. 1—1. Appearance of superconductivity in mercury according to H. Kamerlingh-Onnes (1911). The ordinate is the resistance R;  $R_{\odot}$ , the resistance of solid mercury extrapolated to  $0^{\circ}$  C, is 60 ohms.

which the direct-current resistance reaches one half of the value it had just before the drop is generally given as the transition temperature, because this can be measured accurately. However, a high-frequency investigation to be described in Chap. 16 (f) indicates that the foot of the curve where

<sup>&</sup>lt;sup>1</sup>H. Kamerlingh-Onnes, Commun. Leiden, 120h, 122h, 124c, (1911).

ルーブが疾験し、確認した。と一スナー効果)を保つことを確認

一度競した電影が大なに放れ続

超伝導材料で作ったコイルに

高温品伝導性の可能性を示した された。今春スイスの研究でが 学校 四三郎の「西」で 现金乡、地区国路三〇世(世氏 も起こす新セラミックスが発見 セラミックス 年以此にかぜ口になる可信号 怨人 品構造で、企業のような性質を 一ウムが起じったもの。ベロブス れまで処用化されている担任が カイト型とよばれる三次元の私 も以献しそうた。 担失のない。盗位などの実力化に 動くので、磁気が上列川や電力 た。新材料はヘリウムよらの点 ウムを使わなくてはならなかっ たため、冷却に高価な液体ヘリ いと、昼伝与現象が起きなかっ 材料は超対風医 一〇度以下でな

安価な技体水素で十分

新たに作られたセラミックス

なかった。

程、常作されている意思

の明婚は世界中の研究がかしの

るので、消費で曲く用伝導材料

理工学科の田中昭二教授らのグ 物質で、このほど東大工学部物 一会く迎さない完全反母性(マイ まで、超低吸材料投資の低気を セラミックスが絶対風陰三〇段 持つ。田中研究的は今日、この | やされている。 日本で使われる 一方人(北京総対益度問度)でお 一母質器は、希少物質の液体ヘリ 一般体へリウムは、全帯デメリカ ぎを加っていた。

|はランタンと何の般化物にパリ||録でさえ、三・・・・||控削後で、と |二〇度以下・実験祭での展覧 |の氾禁は一九七三年以来変わら 一致を起こす高い私にが近すさる めて大きい。しかし、科伝母丸 がむられるだと、利用価値は依 ととが火災だった。 プ合から、関が転じは絶対以び すでに実用化されている二十 一伝事材料を使う既粉は増えてい 核田以共和町船舎所数成など起 が可能となる。 |中国二七位の液体ネオンの使用 点が絶対は匹一〇匹の液体水率 際がは度が三〇だになれば、死 などからの輸入に知っている。 和伝染低石、医学用の超低級

asahi shinkun from

> ternational Satellite Edition 1986 (London) 11.

けれいかしているわけじゃあり

観に、整町六ホールの投

いった整備だ。 できるだけ呼く州しておけ 期の判断を迫られた形の気体 国上庁とともに都から飛島時

たという。たて、独切れとか海 間の変色とか。まだ不気味な我る。桶服外のことです」とい 長りというのが、技术的な立場、知を判断しろといわれても、わっち。それらを総合的に判断する だい。<br />
その方向で対策を辿め お続出しているだけに、おい れたは映画できないのが現状 (竹木四)・竹以企団群 に頻繁には聞けないし、帰心時 製作で「子知るの会議もそんな 庁。内田英治及官が弱り切った れわれのできる新聞を超えてい

予知事の簡単は、ある点では 一つの参考ケースがある。昭

町、野境地区、とまでは図って、出たのは、順火から五年後だっ 予知語(安全教育)といる 危険度が低いといえるのが元 いまの状態の中でも、ふくらか さない方針だ。ただし、危険なるものです。学者に責任を特だ すべて多くならないかざり、出 火山活動終息監督は、変色極端、り、逃げる際の点呼などは責任 ア人山性機動などの異常規象が その可能性があることから、 を持ってもらった。住民に責任 機だった。ただし、自治会を作 と、とても町民が収まらないな も持ってもらうと、きちんとも せるのは気の物です」

には宋知子と何対町は

歴化当たっており 記録は一体

じで行われるととにな

住宅準備

確立すれば、安全は増ずたろめている。 ついて回る。また、避難体例を 科学とは別のたくさんの価値が し、帰島には鼠蛇や教育など、 危険といわざるをえない。しか 「科学者としてはいまの状態を **吐火の
危険が
強く
解乱
のめど** 

●有珠山の例 い」というパラパラを練った。 [1] 権に住みた 民の心情をどう両立させるか

次郎さんべうは一枚のブローと、ずっと好きだったから れた大層町数子幣の玉像文(七年に復興して来たけれ 最後の一人、として敷出さ、マイドを手に入れた。二十 伊豆大島から二十七日、見て、ようやく現地でプロ お年寄りがプロマイド

の映画女優、アン・パクス、 の写真について思い出を順「イヴの縁(すべ)で」 立広尾野院のベッドで、そり)の刃」(一九四六年) ターニを第一だった。 マイドを持っていた。米国、村っていた」 玉屋さんは、入院した都一貫を受けた「剃刀(かみそ 作に、アカデミー財政な アン・パクスターは代表 (五〇年) をがあり、昨

女の人も名前さえ知らな代だから、彼女の映画を一 終った。猫の緊張局 えていた昭和二十五年的 い。が、感激した。三度も一番よく見た世代でしょう」 しざに心を打たれた。それ、た人ではないが、知性的な かなにかで、見た女優の美 「関準パラオ諸島に抑留 め、六十二歳で死去した。 んは、「爆発的な人気があっ 年七二月、脳内山血のた 映画評論家の水野時間さ

クスターの思い出格がて 独りぼっち一週間の

変更の子がは、哲学で いまの芸婦一次はいかかって 年度からのスタート。しかし、一向もある。(後 校からは戸惑い、区苑の声があ一まれるのは必至と心 もせっかちな日程だ。大学、高一わったら、新じい序列に 大場等中の旦子にして歌子 を大田様と祖の 関と地大い選手の

た。鉄分など労使協関路級の航

会から応る歴史政立党制は

|上野のホテルで二日間の日程||あいさつした故障組合具は開発 六千人)は二十七日、東京・ 於外心原好通程在、男 と共同歩調も 終始食物

一で、解析状子中央教育を明い

87大きい。 しかし、弱伝導我 | 中向 | 七度の液体ネオンの使用 | 概を取り得る用意がある| と安 が得られるなど、利用価値は極一点が絶対風度一〇度の液体水素一の運動を理解するならば共同後 一が他となる。

金く地を行い完全反映性(マイ)準備的は、帯少物質の液体(リー教授の話・スイスと日本の研究 一・二度前後で、との記録は一九一研究者がしのぎを削っていた。 実験型での最高記録でさえ、三三 | く超伝媒材料の閉究は世界中の 化されているニオブ合金も、四 とが久見った。すでに青 ループ代表、中島長路・東海大 で高温の超伝導が存在すること う機能は地元でおり、西屋で 層所護を超伝統科を 文配省・新雄伝導物質研究グ 医用の軽燥を接続

草協への参加を呼びかけた。

一が、田中教授はセラミックスで一度から六十五年度まで、 場の五方年計画を決事る。此一五十 水道整備などもつの公共等級四一人でといる。 路は天日の間で、第 普及率8%アップ - 水道50カ年計画

我抵抗がゼロになる政団は『物質で、とのほど東大・平都物一たため、冷却に高価な液体へリーと関係してろだ。 新たに作られたセラミックス|スナー効果)を保つことを確解|ウム(形成般対風度四度)で冷 忠づ、超伝導材料特有の破処を セラミックスが絶対孤度三O度 持つ。
田研究家は今月、との

|ループが突動し、硫酸した。と | た。 新材料はヘリウムより書点 | はランタンと何の酸化物にパリ K 担任教材料で作ったコイルに

も知己す析セラミックスが飛晃 | れまで実用化されている超伝導 | が高く、安価な液体水素で十分 | ウムが残じったもの。 ペロブス | 女科は絶対乱医||一〇度19下では | 参くので、磁気浮上列車や電力 | カイト型とよばれる三次元の紀 | 一度能した電流が永久に流れ続

一などからの軽大に掘っている。

臨卵極度が三〇度になれば、赤一の経験が低く有別だ

**題数は源性の可能性を示した一いと、超低導現象が起きなかっ一抵失のない決策などの実用化に「品種造で、金属のような性質を一ければ、少ない電力で強い破場** 

れた。今受くるの政党が

東大・約20mg(行氏 | 35平 科の田中昭 | 教授らのグ | ウムを使わなくてはならなかっ

| 旧主流状が周辺して新型食器 一家職を統一するのが目的、国 として表際、決定して組合の

左尾目に

の運動について動植し、改造 成していることに触れ、こ

有珠山の火山西町町

下韓大輔予知連会長がいう。

は、すでに住宅確保の整備を連 そろいら 事態に備え、東京都 は長期にわたってたたない!

問題は、一万人四半戸といか

のが行政の仕事だら思う」

七三年以来取わらなかった。 異な、青化されている重要

一枚体へリウムは、金サメリカ やされている。日本で使われる

た。今後の眼臓は宗教物のない

#### DISCOVERY OF NEW SUPERCONDUCTING MATERIAL

# " CERAMIC WITH SUFFICIENT SUPERCONDUCTIVE POWER IN HIGH TEMPERATURE REGION "

A new ceramic with a very high  $T_{\rm C}$  of 30K of the superconducting transition has been found. The possibility of high  $T_{\rm C}$  - superconductivity has been reported by scientists in Switzerland in this spring. The group of Prof. Shoji TANAKA, Dept. Appl.Phys. Faculty of Engineering at the University of Tokyo confirmed in November, that this is true.  $T_{\rm C}$ 's of all superconducting materials which we have in practical application till now are lower than 20K. Therefore we need large amount of liquid He for cooling. Note that the price of liquid He is very expensive. But with this new material we can use cheaper liquid  $H_2$  for cooling. We can expect great from this material to the applications such as linear motorcars, electricity transport systems, etc.

The ceramic newly discovered, is an oxide compound of La and Cu with Ba, which has a structure of the so-called perovskite and shows metal-like properties: Prof. Tanaka's laboratory confirmed that this material shows diamagnetism (Meissner effect) up to 30K, which is the most important indication of the existence of superconductivity.

There are a lot of possibilities for practical applications of superconductors. For example very strong magnets, made of superconducting coils, etc. But one handicap is that  $T_C$  is too low in in each material we know till now.

The  $T_{\rm C}$  of Nb-alloys which are already in practical use are lower than 20K. The record of  $T_{\rm C}$  in a laboratory is around 23.2K. This record has not been broken since 1973.

Nowadays each instrument using superconductors is operated by liquid He cooling, and He is a very rare material with a boiling point of 4K. Liquid He used in present Japan is exclusively imported from the USA. If we could get a material with a high T of 30K, we can not only use liquid  $\rm H_2$  but also liquid Ne with a boiling point of 27K.

Since the application of superconductors to many fields, such as very strong magnets, medical use of NMR machines, etc. show rapid increasing, research field of hich  $T_{\rm C}$  superconductivity is high-competitive all over the world.

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## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J. Bednorz et al.

Date: December 18, 1998

Serial No. 08/303,561

Group Art Unit: 1105

Filed: September 9, 1994

Examiner: M. Kopec

For: NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE, AND METHODS FOR THEIR

**USE AND PREPARATION** 

The Commissioner of Patents and Trademarks Washington, D.C. 20231

# **AMENDMENT AFTER FINAL REJECTION UNDER 37 CFR 1.116**

Sir:

In response to the Office Action dated June 25, 1998, please consider the following:

#### **REMARKS**

In the referenced final Office Action, the Examiner refers to three affidavits (of Mitzi, Tsuei and Dinger) submitted by applicants to overcome the rejection under 35 USC 112 for a lack of enablement. The Examiner's comments are at (paragraph (d)(iv) page 7, 5 lines from the bottom to page 8, 4 lines from the bottom. The Examiner states:

(d)(iv)(1) "Those affidavits do not set forth particular facts to support the conclusions that all superconductors based on the applicants' work behave in the same way and that one skilled in the art can make these superconductors without undue experimentation."

(d)(iv)(2) "Those affidavits do not overcome the non-enablement rejection."

(d)(iv)(3) "Those affidavits are not deemed to shed light on the state of the art and enablement at the time the invention was made."

Moreover, in applicant's response dated May 14, 1998, applicants refer to Poole et al. which states at page 59 thereof "[c]opper oxide superconductors with a parity sufficient to exhibit zero resistivity or to demonstrate levitation (Early) are not difficult to snythesize. We believe that this is at least partially responsible for the explosive worldwide growth in these materials" (see applicant's response for the entire text that is quoted and Attachment A thereof for copies of relevant pages from Pool et al.)

In response the Examiner states:

- (1) Initially, however, it should be noted that the Poole article was published *after* the priority date presently claimed. As such, it does not provide evidence of the state of the art *at the time* the presently claimed invention was made.
- (2) Moreover, the present claims are directed to processes of using metal oxide superconductors, not processes of making them. Even if the Poole article provided direct evidence of the state of the art at the time the invention was made, which it apparently does not, that evidence still does not pertain to the issue at hand, namely, the process of using metal oxide superconductors to conduct electricity under superconducting condition.

Applicants respectfully disagree with the Examiner. In further support of applicants position that all their claims are fully enabled, applicant's submit herewith the affidavit under 37 CFR 1.132 of Peter Duncombe which provides a list of books and articles published prior to applicants filing date showing the general principles of ceramic science used by applicants and which are used as stated by Poole et al. to make the

high Tc superconductors taught and claimed by applicants which "are not difficult to synthesize."

The affidavit of Peter Duncombe provides complete copies of two of his notebooks in which he sets forth particular facts in which he recorded the synthesis and properties of high Tc superconducting materials fabricated according to the general principles of ceramic science as taught by applicants.

Applicants submitted by facsimile an affidavit of James W. Leonard on December 15, 1998 which states that 5,689 articles cited the applicants' Zeitschrift fur Physik B-Condensed Matter, **64**, pp. 189-193 (Sept. 1986) article. It cost \$2.50 per citation to print each citation for a total cost of \$14,222.50. Applicants will supply this list at the USPTO's request.

In view of the changes to the claims and the remarks herein, the Examiner is respectfully requested to reconsider the above-identified application. If the Examiner wishes to discuss the application further, or if additional information would be required, the undersigned will cooperate fully to assist in the prosecution of this application.

Please charge any fee necessary to enter this paper to deposit account 09-0468.

If the above-identified Examiner's Action is a final Action, and if the above-identified application will be abandoned without further action by applicants, applicants file a Notice of Appeal to the Board of Appeals and Interferences appealing the final rejection of the claims in the above-identified Examiner's Action. Please charge deposit account 09-0468 any fee necessary to enter such Notice of Appeal.

In the event that this amendment does not result in allowance of all such claims, the undersigned attorney respectfully requests a telephone interview at the Examiner's earliest convenience.

#### MPEP 713.01 states in part as follows:

Where the response to a first complete action includes a request for an interview or a telephone consultation to be initiated by the examiner, ... the examiner, as soon as he or she has considered the effect of the response, should grant such request if it appears that the interview or consultation would result in expediting the case to a final action.

Respectfully submitted,

Daniel P. Morris Reg. No. 32,053

IBM CORPORATION Intellectual Property Law Dept. P.O. Box 218 Yorktown Heights, New York 10598 (914) 945-3217



Applicant:

Bednorz et al.

Art Unit: 1105

Serial No.:

08/303,561

Examiner: D. McGinty

Filed:

September 9, 1994

Date: January 3, 1996

For:

NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIG

TRANSITION TEMPERATURE, AND METHODS FOR THEIR

**USE AND PREPARATION** 

I hereby certify that this paper is being facsimile transmitted under Rule 37 CFR §1.161(d) to the U.S. Patent and Trademark Office on the date shown above.

Daniel P. Morris

Reg. No. 32.053

#### SUPPLEMENTARY RESPONSE

The Commissioner of Patents and Trademarks Washington, D.C. 20231

Sir:

In response to the Office Action dated March 29, 1995, please consider the following:

#### REMARKS

These remarks are in addition to those of the previously submitted response.

As further support for applicants' position that the claims under examination are supported by applicants' specification the attached affidavit of Dr. Chang C. Tsuei is submitted. Dr. Tsuei's affidavit is in agreement with the earlier submitted affidavit of Drs. Donger and Mitzi and states that applicants initi-

ated high temperature superconductor field and the teaching in applicants' specification enables a person of skill in the art to fabricate and use the invention as claimed by applicants.

Claims 24-26, 86-90 and 96-108 have been rejected under 35 USC §102(a) as being anticipated by the Asahi Shinbum article and under 35 USC §103 in view of the Asahi Shinbum article. In addition to applicants' remarks in regard to this rejection in applicant's prior response please consider the following.

The date of the Asahi Shinbum article is November 28, 1986. As stated in applicants' specification at page 6, lines 7-10:

The basis for our invention has been described by us in the following previously published article: J.G. Bednorz and K.A. Muller, Zeitschrift fur Physik B - Condensed Matter, 64, pp. 189-193 Sept. (1986)

The Examiner is using Asahi Shinbum as a reference under 35 USC §102(a). Applicants respectfully disagree since to do so does not permit applicants the one year period provided under 35 USC §102(b) to file a US application after their own publication which permitted applicants to file the present application up to September 1987. The date of the Asahi Shinbum article is after the date of applicants' publication.

Applicants believe this is not a correct application of 35 USC §102. The Court of Custom and Patent Appeal in In re Katz 215 USPQ 14, 17 (a copy of which is attached) states that

It may not be readily apparent from the statutory language that a printed publication cannot stand as a reference under  $\S102(a)$  unless it is describing the work of another. A literal reading might appear to make a prior patent or printed publication "prior art" even though the disclosure is that of the applicant's own work. However, such an interpretation of this section of the statute would negate the one year period afforded under  $\S102(b)^1$  during which an inventor is allowed

to perfect, develop and apply for a patent on his invention and publish descriptions of it if he wishes.

Thus, one's own work is not prior art under §102(a) even though it has been disclosed to the public in a manner or form which otherwise would fall under §102(a). Disclosure to the public of one's own work constitutes a bar to the grant of a patent claiming the subject matter obvious therefrom only when the disclosure occurred more than one year prior to the date of the application, that is, when the disclosure creates a one-year time bar, frequently termed a "statutory bar," to the application under §102(b). As stated by this court in In re Facius, 56 CCPA 1348, 1358, 408 F.2d 1396, 1406, 161 USPQ 294, 302 (1969), "But certainly one's own invention, whatever the form of disclosure to the public, may not be prior art against oneself, absent a statutory bar." [Emphasis in original]<sup>2</sup>.

The Asahi Shinbum article states in the first paragraph:

A new ceramic with a very high  $T_c$  of 30K of the superconducting transition has been found. The possibility of high  $T_c$  -superconductivity has been reported by scientists in Switzerland this spring. The group of Prof. Shoji TANAKA, Dept. Appl. Phy. Faculty of Engineering at the University of Tokyo confirmed in November, that this is true.

The "scientists in Switzerland" are the inventors of the above-identified application. The Asahi Shinbum article only reports the work of applicants and that it was reproduced by Prof. Tanaka. This article is a disclosure of applicants' "own invention" and cannot be used as a reference. Therefore, the Examiner is respectfully requested to withdraw the rejection of claims 24-26, 86-90 and 96-108 under 35 USC §102(a) as anticipated by Asahi Shinbum and under 35 USC §103 as obvious over Asahi Shinbum.

Daniel P. Morris Registration No. 32,053



#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:

Bednorz et al.

Art Unit: 1105

Serial No.:

08/303,561

Examiner: D. McGinty

Filed:

September 9, 1994

Date: January 2, 1996

For:

NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE, AND METHODS FOR THEIR

**USE AND PREPARATION** 

#### AFFIDAVIT UNDER 37 C.F.R. §1.132

The Commissioner of Patents and Trademarks Washington, D.C. 20231

Sir:

I, Chang C. Tsuei, being duly sworn, do hereby depose and state:

That I received a B.S. degree in Mechanical Engineering from National Taiwan University (1960) and M.S. and PhD. degrees, in Material Science (1963, 1966) respectively from California Institute of Technology.

That I have worked as a research staff member and manager in the physics of superconducting, amorphous and structured materials at the Thomas Watson Research Center of the International Business Machines Corporation in Yorktown Heights, New York, from 1973 to the present. (See attached Exhibit A for other professional employment history.)

That I have worked in the fabrication of and characterization of high temperature superconductor and related materials from 1973 to the present.

That I have reviewed the above-identified patent application and acknowledge that it represents the work of Bednorz and Muller, which is generally recognized as the first discovery of superconductivity above 26°K and that subsequent developments in this field have been based on this work.

That all the high temperature superconductors which have been developed based on the work of Bednorz and Muller behave in a similar manner, conduct current in a similar manner and have similar magnetic properties.

That once a person of skill in the art knows of a specific transition metal oxide composition which is superconducting above 26°K, such a person of skill in the art, using the techniques described in the above-identified patent application, which includes all known principles of ceramic fabrication, can make the transition metal oxide compositions encompassed by claims 24-26, 86-90 and 96-108, without undue experimentation or without requiring ingenuity beyond that expected of a person of skill in the art. This is why the work of Bednorz and Muller was reproduced so quickly after their discovery and why so much additional work was done in this field within a short period of their discovery.

By: Chyc. Tr

Chang C. Tsuei

Sworn to before me this 26th day of September, 1995.

Notary Public

BANIEL P. MORRIS NOTARY PUBLIC, State of New York

No. 4838676
Qualified in Westchester County
Commission Expires March 15, 19.7.7

#### **CHANG C. TSUEI**

#### Education

California Institute of Technology, M.S. (1963), Ph.D. (1966) National Taiwan University, B.S. (1960)

#### **Professional Employment**

1993-present - Research Staff Member

1983-1993 - Manager, Physics of Structured Materials

1979-1983 - Manager, Physics of Amorphous Materials

1974-1975 - Acting Manager, Superconductivity

1973-1979 - Research Staff Member

Harvard University: 1980 (Summer)

Visiting Scholar in Applied Physics

Stanford University: 1982 (Sept.) - 1983 (April)

Visiting Scholar in Applied Physics

#### California Institute of Technology

1972 - 1973 - Senior Research Associate in Applied Physics

1969 - 1972 - Senior Research Fellow in Materials Science

1966 - 1969 - Research Fellow in Materials Science

#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE



Applicants: J.G. BEDNORZ ET AL.

: Date: March 21, 1988

Filed: 05/22/87

: Serial No.: 06/053,307

Group Art Unit: 115

: Examiner: Dennis Albrecht

FOR: NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE,

AND METHODS FOR THEIR USE AND PREPARATION

DECLARATION OF J. GEORG BEDNORZ AND K.A. MUELLER WITH RESPECT TO HIGH To SUPERCONDUCTIVITY

Commissioner of Patents and Trademarks Washington, D. C. 20231

Sir:

We, J. Georg Bednorz and K.A. Mueller, hereinafter say and declare the following:

1. We are the inventors of the contribution described and claimed in the subject U.S. patent application. This application describes our earlier discovery of high temperature superconductivity in ceramic copper oxide materials. In particular, one of the earlier systems in which we worked was comprised of Ba-La-Cu-oxides which exhibited superconducting onset temperatures in the mid unter thirty K range.



- 2. We conducted extensive research on these materials to establish their superconductivity in our laboratory in Zurich, Switzer- land. This work traced back to early 1986 and was described in a publication by us that appeared in Z. Phys. B - Condensed Matter 64, 189-193 (1986). This article describes, among other items, resistivity versus temperature measurements that we made on samples of this material to show its superconductive behavior. We subsequently submitted for publication and had published additional articles describing these high Tc oxide superconductors.
- 3. On approximately October 16, 1986, we gave Praveen Chaudhari (Vice-President, Science at the Yorktown Research Laboratory of IBM Corporation) six samples of the high temperature superconductive ceramic oxide material that we had described in our aforementioned Z. Physik B. publication. Praveen Chaudhari brought these samples back to the U.S. when he returned after

visiting with us on or about October 16, 1986. These samples were given to him so that experimentation and measurement could be performed on the samples in the United States. We knew the individuals (Richard Greene and Chang C. Tsuei) who would be involved in the measurements in the United States and discussed the measurements with these individuals in approximately the third week of October, 1986. We maintained telephone and computer communications with these individuals from that time continually through the remainder of 1986 and into 1987.

- 4. It was decided by us that Richard Greene would do specific heat measurements on these samples while magnetic measurements would be done by us in our Zurich laboratory. Greene worked for Chang Tsuei and discussed with him the nature of the experiments and development activities to be performed at the aforementioned Yorktown lab. We provided guidance to Richard Greene and Chang Tsuei by describing the nature of these superconducting samples and the types of properties that we had measured relative to these samples. One of us (K.A. Mueller) also discussed confirmation of our resistivity versus temperature measurements with said Chang Tsuei in a telephone converation in October, 1986.
- 5. The early work conducted by the individuals in the Yorktown laboratory on our superconducting samples occurred with the supervision and guidance that we furnished to these Yorktown scientists. Addition-ally, we provided a preprint to Richard Greene of an article that we subsequently published in Europhysics Letters 3, (3), pp. 379-385 (1987). This article was given to Greene in October, 1986 and described magnetic measurements on these superconducting samples.
- 6. We were aware of the work being conducted on our samples at the Yorktown lab and were in contact with the individuals there, and particularly Richard Greene, who reported to Chang Tsuei. Since the specif-ic heat measurements generally take longer, we had considerable interactions with Rick Green over a period of time from about October 22, 1986 through December, 1986, concerning his specific heat measurements. We also were made aware of Chang Tsuei's measurements of resistivity versus temperature of the Ba-La-Cu-oxide samples, which confirmed our earlier resistivity versus temperature measurements.
- 7. We further declare that all statements made hereinabove based on our own knowledge are true and that all statements made on information and belief are believed to be true. We further declare

that these statements are made with the knowledge that willful false state- ments and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of our Patent Application or any patent issuing thereon.

J. GEORG BEDNORZ DATE: May 30. 1988

K.A.Cex Muller

K.A. MUELLER

DATE: My 27. 1988



#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J.G. BEDNORZ ET AL. : Date: March , 1988

Filed: 05/22/87 : Serial No.: 06/053,307

Group Art Unit: 115 : Examiner: Dennis Albrecht

FOR: NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE,

AND METHODS FOR THEIR USE AND PREPARATION

# DECLARATION OF ALEXIS P. MALOZEMOFF WITH RESPECT TO HIGH TO SUPERCONDUCTIVITY

Commissioner of Patents and Trademarks Washington, D.C. 20231

#### Sir:

- I, Alexis P. Malozemoff, hereby declare and say that:
- 1. I have a PhD from Stanford University and was a Senior Manager in the Physical Sciences Department at the Thomas J. Watson Research Center of IBM Corporation at Yorktown, N.Y. in the fall of 1986. At that time, Chang C. Tsuei reported to me while Richard Greene and Sung Il Park reported to Chang Tsuei. I had general responsibility for a research program in superconductivity and amorphous materials.
- 2. In approximately September October 1986, I was made aware of the pioneering work in superconductivity done by Bednorz and Mueller in Zurich. I had seen an activity report prepared by the Zurich Research lab of IBM Corporation, which detailed that work and the measurements that Bednorz and Mueller had made on Ba-La-Cu-O ceramic superconductors. In response, I discussed specific heat measurements that could be made on these superconducting materials to complement the work being done at Zurich by Bednorz and Mueller. My discussion was with Richard Greene whom I encouraged to be involved in this technical activity.
- 3. On or about November 15, 1986, Richard Greene and I traveled to Baltimore for a Magnetism conference. During our travel to Baltimore, we discussed Greene's ongoing experiments on high  $T_{\rm C}$  superconducting samples

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which he said had been received from Bednorz and Mueller. Specifically, we discussed Greene's preliminary data on specific heat measurements. Richard Greene reported to me that the diamagnetic signal was present but very small. I encouraged him to continue with his measurements with the anticipation that he might be able to present a paper at the Materials Research Society meeting that was scheduled for December, 1986.

- 4. I recall Chang Tsuei telling me his measurements of resistivity versus temperature on the superconducting samples of Ba-La-Cu-Oxides which had been obtained from Zurich. These measurements were done at the aforementioned Research Center and Tsuei reported that his measurements confirmed earlier measurements of Bednorz and Mueller and were consistent with the results published by Bednorz and Mueller in Z. Phys. B-Condensed Matter 64, pp. 189-193 (1986). My recollection of the exact date Chang Tsuei told me of his resistivity versus temperature measurements is not clear. However, I do know that he told me in either November or December of 1986. To the best of my recollection, Tsuei's discussion with me was shortly after the Materials Research Society meeting in Boston the first week of December, 1986.
- 5. I do have a strong recollection of the work of Richard Greene on the specific heat measurements of these samples and of his measurements of resistivity versus temperature in the presence of a magnetic field. Greene's work at the Research Center started soon after he received the superconducting samples in October, 1986 and continued on a daily basis throughout the remainder of 1986 and into 1987. He discussed with me and showed me data concerning these measurements, which indicated to me that a portion of the samples was superconducting. During this time he told me that he also communicated regularly with Bednorz and Mueller in Zurich to inform them of his work and to coordinate his efforts with those of Bednorz and Mueller. He said that he received technical guidance and support from Bednorz and Mueller and worked in complete collaboration with these individuals.
- 6. I further declare that all statements made hereinabove are of my own knowledge and are true and that all statements made on information and belief are believed by me to be true. Further, I declare that these statements were made in the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under

Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of a Patent Application or any patent issuing thereon.

Clexic & Malozemost DATE: Man. 30, 1988

ALEXIS P. MALOZEMOST /



#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J.G. BEDNORZ ET AL.

: Date: March 29, 1988

Filed: 05/22/87

Serial No.: 06/053,307

Group Art Unit: 115

: Examiner: Dennis Albrecht

FOR: NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE,

AND METHODS FOR THEIR USE AND PREPARATION

#### DECLARATION OF CHENG-CHUNG JOHN CHI WITH RESPECT TO HIGH TC SUPERCONDUCTIVITY

Commissioner of Patents and Trademarks Washington, D. C. 20231

Sir:

- I, Cheng-Chung John Chi, hereby declare and say the following:
- 1. I have a PhD in Physics which I received from the University of Pennsylvania in 1976. After graduation, I did Post-doctoral work at the University of California, Berkeley and then joined the Research Division of IBM Corporation in 1979. I am presently a research staff member on the technical staff of the Director of Research at the Thomas J. Watson Research Center of IBM Corporation located at Yorktown, New York.
- 2. At a time prior to approximately the middle of November, 1986, Chang C. Tsuei told me of measurements he made on samples of high T<sub>C</sub> superconducting material which he said were received from Georg Bednorz and K. A. Mueller, two physicists working for IBM Corporation in Zurich, Switzerland. These samples of superconducting material were La-Ba-Cu-O crystalline materials of the type described by Bednorz and Mueller in Z. Phys. B-Condensed Matter 64, pp. 189-193 (1986). Chang Tsuei said that he had measured resistivity versus temperature of these samples.
- 3. In the time frame mentioned hereinabove, Chang Tsuei showed me plots of resistivity versus temperature for the measurements he had made on these superconducting samples. I recall him telling me that the superconducting transition was "not really sharp", which is to

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be expected for samples prepared during the infancy of a technology development. I recognized that these plots were evidence of superconductivity with high  $\mathbf{T}_{\mathbf{C}}$ , even if the transition were not very sharp at that time. Based on Tsuei's statements to me, I believe that these resistivity versus temperature plots were shown to me within a day or two of the time Tsuei made these measurements in his laboratory.

- 4. All acts described by me in this declaration occurred in the United States.
- 5. I further declare that all statements made hereinabove are of my own knowledge and are true and that all statements made on information and belief are believed by me to be true. Further, I declare that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of a Patent Application or any patent issuing thereon.

Change Chung John Chi

DATE: 3/30/88



#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J.G. BEDNORZ ET AL. : Date: March 29, 1988

Filed: 05/22/87 : Serial No.: 06/053,307

Group Art Unit: 115 : Examiner: Dennis Albrecht

FOR: NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE,

AND METHODS FOR THEIR USE AND PREPARATION

### DECLARATION OF SUNG IL PARK WITH RESPECT TO HIGH TO SUPERCONDUCTIVITY

Commissioner of Patents and Trademarks Washington, D.C. 20231

Sir:

- I, Sung Il Park, hereby declare and say that:
- 1. I received a PhD in Applied Physics from Stanford University in October, 1986. I joined the Thomas J. Watson Research Center of IBM Corporation in Yorktown, N.Y. on approximately October 20, 1986, and began work as a Post-doctoral employee. My initial assignment was in the areas of Thin Film Interfaces and high  $T_{\rm C}$  superconductivity. Almost from the beginning of my employment by IBM Corporation I worked exclusively on high  $T_{\rm C}$  superconductivity and reported to Chang C. Tsuei. I am presently working in the same capacity for Chang C. Tsuei, one hundred percent of my time being spent on high  $T_{\rm C}$  superconductivity.
- 2. I was told by Chang C. Tsuei that superconducting samples had been received by Richard Greene, the samples having been brought from IBM Corporation's Research Lab in Zurich, Switzerland. These were said to be Ba-La-Cu-Oxides that had been fabricated by Georg Bednorz and Alex Mueller in Zurich, Switzerland and transported to the U.S. by Praveen Chaudhari. Soon after I began working for Chang C. Tsuei, he asked me to prepare two of these samples for measurements of resistivity versus temperature. To do so, I and Greene cut pieces from these samples to be used for the measurements. I then pressed indium dots into these cut samples to provide electrical contacts. I attached copper wires

to the indium dots in order to allow connections to a current source and to voltage-measuring equipment. The individual copper wires were given number designations to allow them to be properly attached to the equipment used for the current and voltage measurements. This numbering system is represented by the two figures appearing in Chang C. Tsuei's laboratory notebook, a true copy of two pages of which are attached hereto and labeled Exhibit A.

- In order to determine resistivity versus temperature, measurements were made of the current flowing through the Ba-La-Cu-Oxide sample while the voltage across two of the copper leads was measured. Both positive and negative polarity currents were used in order to avoid thermal effects that sometimes occur when making DC measurements. superconducting sample was located on the end of a long probe and placed in a dewar containing liquid helium. The temperature was varied by using a heater placed near the sample. Data of current and voltage were taken from 4.2K to 50K. A germanium thermometer near the sample was used to measure the sample temperature. Since resistance is proportional to voltage, the voltage and current measurements allowed the resistance (and therefore the resistivity for a sample of known dimensions) to be measured as a function of temperature. I worked with Chang C. Tsuei to take these measurements and used a xy recorder to provide graphical plots of resistivity versus temperature for the temperature range 50K-4.2K for at least two of these Ba-La-Cu-Oxide samples.
- 4. The preparation and measurement of the aforementioned superconducting samples occurred at a date prior to November 15, 1986, and to the best of my recollection occurred on or about November 9, 1986, the date when a helium dewar was pumped down preparatory to taking the actual measurements. I believe that while I was assisting Chang C. Tsuei and working under his direction, Bradford Orr observed our data and graphical plots, and we told him the nature of the superconducting samples and the types of measurements that we were making.
- 5. My recollection of the dates when the preparation and measurement of these samples occurred is vivid to me. My first week of employment under Chang C. Tsuei was spent looking for an apartment and, upon beginning laboratory work the following week (about October 28, 1986), I was instructed by Chang C. Tsuei to immediately measure the

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aforementioned samples. I cut pieces from these samples using a fine wire cutter following the direction of Richard Greene. This was done on or about October 28, 1986. After this I contacted the samples with indium as described above. This technique was very familiar to me as I had used indium contacts many times at Stanford University.

- 6. Both Chang C. Tsuei and I were enthused about our measurement results, as the data showed a superconducting onset temperature of about 35K followed by a drop to zero DC resistivity. We noted that the transition to zero resistivity was fairly wide, which we expected to be the case for samples that may have been unperfected and not of a single phase. In fact, the question of whether multiple phases were present in the material was something that was noted by Tsuei in the aforementioned laboratory notebook, identified as Exhibit A, attached hereto.
- 7. At this time, I have been unable to locate the actual data and graphical plots of resistivity versus temperature described hereinabove. I believe that this data may have been misplaced or inadvertently thrown out during an extensive cleaning of the laboratory. However, my memory of the events preparing the samples for measurements, performing the measurements and recording the data, and the results indicated by that data, are very clear in my mind.
- 8. All of the acts described in paragraphs 1 7 above occurred in the United States.
- 9. I further declare that all statements made hereinabove are of my own knowledge and are true and that all statements made on information and belief are believed by me to be true. Further, I declare that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of a Patent Application or any patent issuing thereon.

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#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J.G. BEDNORZ ET AL.

: Date: March 29, 1988

Filed: 05/22/87

: Serial No.: 06/053,307

Group Art Unit: 115

Examiner: Dennis Albrecht

FOR: NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE,

AND METHODS FOR THEIR USE AND PREPARATION

#### DECLARATION OF CHANG C. TSUEI

#### WITH RESPECT TO HIGH To SUPERCONDUCTIVITY

Commissioner of Patents and Trademarks Washington, D.C. 20231

Sir:

- I, Chang C. Tsuei, hereby declare and say that:
- 1. I have a PhD in Material Science from California Institute of Technology, and worked approximately eleven years as a student and faculty member at Cal. Tech. During this time, my research was primarily on amorphous materials and superconducting materials. I joined the Thomas J. Watson Research Center of IBM Corporation in Yorktown, N.Y. in 1973 and continued my work on amorphous materials. For several years, I have been the Manager of a group in the Physical Sciences Department studying amorphous superconductivity and superconductivity of new High  $\mathbf{T}_{\mathbf{C}}$  superconducting ceramic materials.
- 2. On approximately September 13, 1986, I returned from a sabbatical at the K. Onnes Laboratory in Holland. Upon my return, I saw a copy of an IBM activity report for May June, 1986, in which the "novel research" of J.G. Bednorz and K.A. Mueller was described. These individuals were working in the Zurich, Switzerland research laboratory of IBM Corporation, and had observed a steep decrease of resistivity in sintered samples of Ba-La-Cu-oxides. A true copy of this activity report is attached hereto and labeled Exhibit A. On pasge 2 of the activity report the resistivity versus temperature plot is shown in Figure 1

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wherein the onset temperature for superconductivity is in the 35K range. The data and measurements discussed in the activity report were later published by Bednorz and Mueller in Z. Phys. B-Condensed Matter, 64, pp. 189 - 193 (1986), a true copy of which is attached and labeled Exhibit B. Based on my previous experience in superconductivity, I was very interested in the work of Bednorz and Mueller and discussed this work with my colleague, Richard Greene (who reported to me). I told Greene to review this activity report and to start a project on high  $T_{\rm C}$  superconductors of the type described by Bednorz and Mueller. This project was started by Richard Greene and others in the group that reported to me, almost immediately.

- 3. I called Alex Mueller in Switzerland via telephone to request samples of his superconducting material, as well as to discuss the technical area with him. I also sent computer messages to Mueller, but could not contact him. After this, early in October, 1986, I obtained a copy of the aforementioned Z. Phys. B article by Bednorz and Mueller.
- I knew that on approximately October 17, 1986, Praveen Chaudhari was in Zurich, Switzerland. I was told that he was given samples of the Bednorz and Mueller superconducting copper oxides to bring to the U.S. for collaborative work in the United States. I was also told that these superconducting samples were delivered to Richard Greene on or about October 22, 1986. Shortly after these superconducting samples were received, I began work to confirm the existence of high temperature superconductivity in these materials and instructed Sung Il Park to assist me. To do so, small pieces of these samples were cut by Park and Greene and were prepared with indium contact dots to which copper wires were attached. These copper wires were attached to a source of electrical current and to voltage-measuring equipment to determine the existence of the superconducting state. As the temperature of the sample was lowered, resistivity versus temperature plots were then made using standard laboratory techniques. The preparation of these samples for measurement was done by Sung Il Park, who reported to me and who was directed by me to do so.
- 5. A true copy of the cover sheet and a page of my laboratory notebook is attached hereto and labeled Exhibit C. On the second page of

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this exhibit, two diagrams illustrate the samples and show the location of the indium contact dots on the superconducting samples and the numbering given to the copper wires attached to these dots. This numbering enabled us to properly connect these wires to a current source and to voltage-measuring equipment. All entries on this notebook page were made by me. The samples are generally designated by their composition, the term "BLCO" standing for Ba-La-Cu-Oxide materials. The designation "BLCO 21 -- II," etc. in the box in the right hand corner of this page and the designation "BLCO 2 I" in the circle in the middle of the page were the designations on the sample boxes in which the samples were located. copied these designations directly into my notebook. On the bottom right hand corner of this exhibit, the words "dewar pumped  $3 imes 10^{-5}$  Torr 11/9/86" is indicated. I made this notation on November 9, 1986 indicating that the dewar was being pumped down in order to enable the resistivity versus temperature measurements to be made. Because this dewar leaked, the actual measurements had to be made within several hours of the pump-down.

- 6. The individual superconducting samples were attached to a long probe and slowly lowered into the liquid helium dewar while a current was passed through the sample and the voltage across two of the terminals measured. Sung Il Park assisted me. These measurements were recorded directly on an xy recorder which plotted resistivity versus temperature for these superconducting samples. These plots indicated an onset of superconductivity at about approximately 35K, and confirmed the results of Bednorz and Mueller in Switzerland. As an example, referring to the sample BLCO 2 having connecting wires 20, 21, 22 and 8, electrical current was applied between wires 20 and 8, while voltage measurements were made across the sample using contact terminals 21 and 22. voltage is a function of the resistance of the material, by making the voltage measurements at constant currents, resistivity versus temperature plots can be developed. These resistivity versus temperature plots appear to be missing at this time. I believe that they may have been inadvertently thrown away when the laboratory was subsequently extensively cleaned.
- 7. During my measurement of the superconducting samples described hereinabove, Bradford G. Orr, who was a Post-doctoral employee

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at the Research Center, came into my lab and observed these measurements, as did Albert M. Torressen. These measurements confirmed the high temperature superconductivity of these materials and I was enthusiastic about the results. I expressed my enthusiasm to Richard Greene, who was anxious to do specific heat measurements on these samples. Subsequent to my confirmation of their resistivity versus temperature measurements, I contacted Mueller in Zurich to inform him of my successful resistivity versus temperature measurements. In addition, I told several people about my laboratory measurements, including Arthur Williams, Alex Malozemoff, Paul Horn, and Praveen Chaudhari, all of whom work in the Thomas J. Watson Research Center.

- 8. From the time we received the superconducting samples in October, 1986 to the present, I, Richard Greene and Sung Il Park have worked on a daily basis to further explore and develop this technology. In particular, I observed Greene working on a daily basis to conduct specific heat measurements during November and December, 1986.
- 9. All acts described hereinabove relating to sample preparation, measurement and discussions of these measurements occurred in the United States.
- 10. I further declare that all statements made hereinabove are of my own knowledge and are true and that all statements made on information and belief are believed by me to be true. Further, I declare that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of a Patent Application or any patent issuing thereon.

Chang C. Tsuei

DATE: 3/30/88

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Zurich Research Laboratory 8803 Rüschlikon, Switzerland

Telephone: \$1 724 81 11

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IBM

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ACTIVITY REPORT MAY-JUNE, 1986 August 15; 1986

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**MATERIAL SCIENCE** 

T. Schneider, Mgr.

SURFACE & MATERIAL SCIENCES E. Courtens, Mgr., Project 4181

**Novel Research** 

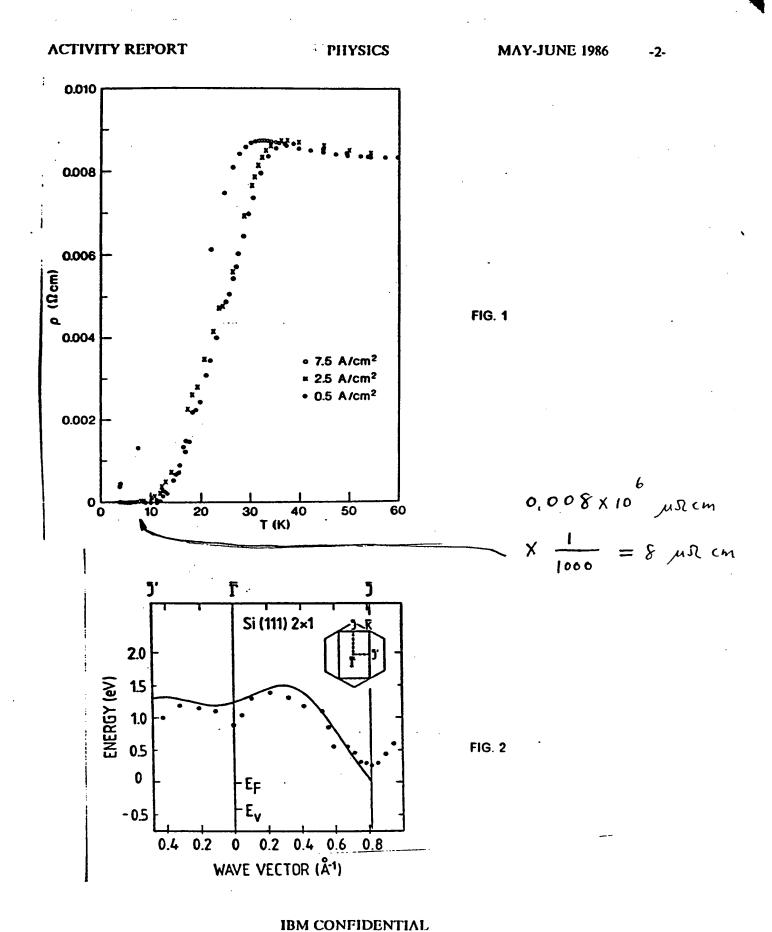
Possible High-T, Superconductivity in the Ba-La-Cu-O System

J.G. Bednorz and K.A. Müller (Project 4196)

We observed a steep decrease of resistivity in sintered Ba-La-Cu-oxide samples, with the highest temperature of the onset in the 35 K range (Fig. 1).

The Ba-La-Cu-O system exhibits a number of oxygen deficient phases with perovskite-like layer-type structures. These are characterized by mixed-valent copper ions (Cu<sup>2+</sup> and Cu<sup>3+</sup>) and itinerant electronic states. In addition one expects polaron formation induced by the strong Jahn-Teller effect of Cu<sup>2+</sup> in an octahedral oxygen environment. Thus our Ba-La-Cu-O system was anticipated to have considerable electron-phonon coupling and metallic conductivity.

Compounds with the composition Ba(x)La(5-x)Cu(5)O(5[3-y]) have been prepared in polycrystalline form. Samples with x < 0.2 and y > 0, annealed below 900°C under reducing conditions, consist of three phases, one of them a perovskite-like mixed-valent copper compound with  $K_2NiF_4$  type structure. Upon cooling, the samples show a linear decrease in resistivity, then an approximately logarithmic increase, interpreted as a beginning of localization. Finally a steep decrease by up to three orders of magnitude occurs, reminiscent of the onset of percolative superconductivity. The highest onset temperature is observed in the 35 K range. It is markedly reduced by high current densities (Fig. 1). The slow sensitivity decay towards low temperatures might possibly result from 2D superconducting fluctuations of perovskite layers of one of the phases present.



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# Possible High $T_c$ Superconductivity in the Ba-La-Cu-O System

J.G. Bednorz and K.A. Müller
IBM Zürich Research Laboratory, Rüschlikon, Switzerland

Received April 17, 1986

Metallic, oxygen-deficient compounds in the Ba-La-Cu-O system, with the composition  $Ba_xLa_{5-x}Cu_5O_{5(3-y)}$  have been prepared in polycrystalline form. Samples with x=1 and 0.75, y>0, annealed below 900 °C under reducing conditions, consist of three phases, one of them a perovskite-like mixed-valent copper compound. Upon cooling, the samples show a linear decrease in resistivity, then an approximately logarithmic increase, interpreted as a beginning of localization. Finally an abrupt decrease by up to three orders of magnitude occurs, reminiscent of the onset of percolative superconductivity. The highest onset temperature is observed in the 30 K range. It is markedly reduced by high current densities. Thus, it results partially from the percolative nature, bute possibly also from 2D superconducting fluctuations of double perovskite layers of one of the phases present.

#### I. Introduction

"At the extreme forefront of research in superconductivity is the empirical search for new materials" [1]. Transition-metal alloy compounds of A15 (Nb<sub>3</sub>Sn) and B1· (NbN) structure have so far shown the highest superconducting transition temperatures. Among many A15 compounds, careful optimization of Nb—Ge thin films near the stoichiometric composition of Nb<sub>3</sub>Ge by Gavalev et al. and Testardi et al. a decade ago allowed them to reach the highest  $T_c$  = 23.3 K reported until now [2, 3]. The heavy Fermion systems with low Fermi energy, newly discovered, are not expected to reach very high  $T_c$ 's [4].

Only a small number of oxides is known to exhibit superconductivity. High-temperature superconductivity in the Li-Ti-O system with onsets as high as 13.7 K was reported by Johnston et al. [5]. Their x-ray analysis revealed the presence of three different crystallographic phases, one of them, with a spinel structure, showing the high  $T_c$  [5]. Other oxides like perovskites exhibit superconductivity despite their small carrier concentrations, n. In Nb-doped SrTiO<sub>3</sub>, with  $n=2\times10^{20}$  cm<sup>-3</sup>, the plasma edge is below the highest optical phonon, which is therefore unshielded

[6]. This large electron-phonon coupling allows a  $T_c$ of 0.7 K [7] with Cooper pairing. The occurrence of high electron-phonon coupling in another metallic oxide, also a perovskite, became evident with the discovery of superconductivity in the mixed-valent compound BaPb<sub>1-x</sub>Bi<sub>x</sub>O<sub>3</sub> by Sleight et al., also a decade ago [8]. The highest  $T_c$  in homogeneous oxygen-deficient mixed crystals is 13 K with a comparatively low concentration of carries  $n = 2-4 \times 10^{21}$  cm<sup>-3</sup> [9]. Flat electronic bands and a strong breathing mode with a phonon feature near 100 cm<sup>-1</sup>, whose intensity is proportional to  $T_c$ , exist [10]. This last example indicates that within the BCS mechanism, one may find still higher  $T_c$ 's in perovskite-type or related metallic oxides, if the electron-phonon interactions and the carrier densities at the Fermi level can be enhanced further.

Strong electron-phonon interactions in oxides can occur owing to polaron formation as well as in mixed-valent systems. A superconductivity (metallic) to bipolaronic (insulator) transition phase diagram was proposed theoretically by Chakraverty [11]. A mechanism for polaron formation is the Jahn-Teller effect, as studied by Höck et al. [12]. Isolated Fe<sup>4+</sup>, Ni<sup>3+</sup> and Cu<sup>2+</sup> in octahedral oxygen environment

show strong Jahn-Teller (J.T.) effects [13]. While SrFe(VI)O<sub>3</sub> is distorted perovskite insulator, LaNi(III)O<sub>3</sub> is a J.T. undistorted metal in which the transfer energy  $b_{\pi}$  of the J.T.  $e_{\pi}$  electrons is sufficiently large [14] to quench the J.T. distortion. In analogy to Chakraverty's phase diagram, a J.T.-type polaron formation may therefore be expected at the borderline of the metal-insulator transition in mixed perovskites, a subject on which we have recently carried out a series of investigations [15]. Here, we report on the synthesis and electrical measurements of compounds within the Ba-La-Cu-O system. This system exhibits a number of oxygen-deficient phases with mixed-valent copper constituents [16], i.e., with itinerant electronic states between the non-J.T. Cu3+ and the J.T. Cu2+ ions, and thus was expected to have considerable electron-phonon coupling and metallic conductivity.

#### II. Experimental

#### 1. Sample Preparation and Characterization

Samples were prepared by a coprecipitation method from aqueous solutions [17] of Ba-, La- and Cu-nitrate (SPECPURE JMC) in their appropriate ratios. When added to an aqueous solution of oxalic acid as the precipitant, an intimate mixture of the corresponding oxalates was formed. The decomposition of the precipitate and the solid-state reaction were performed by heating at 900 °C for 5 h. The product was pressed into pellets at 4 kbar, and reheated to 900 °C for sintering.

#### 2. X-Ray Analysis

X-ray powder diffractograms (System D 500 SIE-MENS) revealed three individual crystallographic phases. Within a range of 10° to 80° (2 $\theta$ ), 17 lines could be identified to correspond to a layer-type perovskite-like phase, related to the K<sub>2</sub>NiF<sub>4</sub> structure (a=3.79 Å and c=13.21 Å) [16]. The second phase is most probably a cubic one, whose presence depends on the Ba concentration, as the line intensity decreases for smaller x(Ba). The amount of the third phase (volume fraction > 30% from the x-ray intensities) seems to be independent of the starting composition, and shows thermal stability up to 1,000 °C. For higher temperatures, this phase disappears progressively, giving rise to the formation of an oxygen-delicient perovskite (La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14</sub>) as described by Michel and Raveau [16].

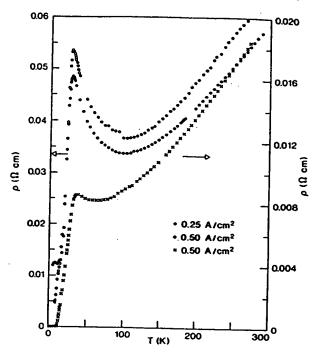


Fig. 1. Temperature dependence of resistivity in  $Ba_xLa_{5-x}Cu_5O_{5(3-y)}$  for samples with x(Ba)=1 (upper curves, left scale) and x(Ba)=0.75 (lower curve, right scale). The first two cases also show the influence of current density

#### 3. Conductivity Measurements

The dc conductivity was measured by the four-point method. Rectangular-shaped samples, cut from the sintered pellets, were provided with gold electrodes and contacted by In wires. Our measurements between 300 and 4.2 K were performed in a continuous-flow cryostat (Leybold-Hereaus) incorporated in a computer-controlled (IBM-PC) fully-automatic system for temperature variation, data acquisition and processing.

For samples with  $x(Ba) \le 1.0$ , the conductivity measurements, involving typical current densities of 0.5 A/cm<sup>2</sup>, generally exhibit a high-temperature metallic behaviour with an increase in resistivity at low temperatures (Fig. 1). At still lower temperatures, a sharp drop in resistivity (>90%) occurs, which for higher currents becomes partially suppressed (Fig. 1: upper curves, left scale). This characteristic drop has been studied as a function of annealing conditions, i.e., temperature and O<sub>2</sub> partial pressure (Fig. 2). For samples annealed in air, the transition from itinerant to localized behaviour, as indicated by the minimum in resistivity in the 80 K range, is not very pronounced. Annealing in a slightly reducing atmosphere, however, leads to an increase in resistivity and a more pronounced localization effect. At the same time, the onset of the resistivity drop is shifted

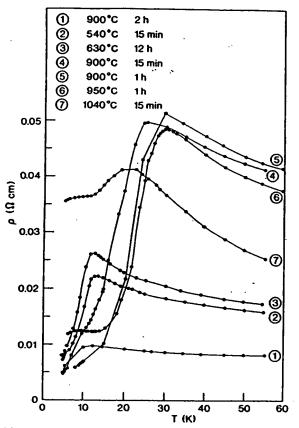


Fig. 2. Low-temperature resistivity of samples with x(Ba) = 1.0, annealed at  $O_2$  partial pressure of 0.2 bar (curve ①) and  $0.2 \times 10^{-4}$  bar (curves ② to ⑦)

towards the 30 K region. Curves (and (3), recorded for samples treated at 900 °C, show the occurrence of a shoulder at still lower temperature, more pronounced in curve 6. At annealing temperatures of 1,040 °C, the highly conducting phase has almost vanished. As mentioned in the Introduction, the mixed-valent state of copper is of importance for electron-phonon coupling. Therefore, the concentration of electrons was varied by the Ba/La ratio. A typical curve for a sample with a lower Ba concentration of 0.75 is shown in Fig. 1 (right scale). Its resistivity decreases by at least three orders of magnitude, giving evidence for the bulk being superconducting below 13 K with an onset around 35 K, as shown in Fig. 3. on an expanded temperature scale. The latter figure also shows the influence of the current density, typical for granular compounds.

#### III. Discussion

The resistivity behaviour of our samples, Fig. 1, is qualitatively very similar to the one reported in the Li-Ti-O system, and in superconducting

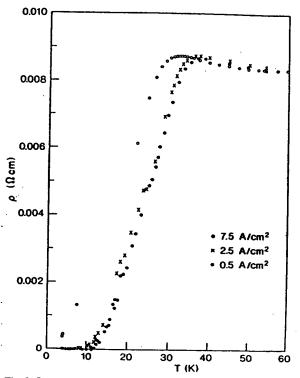


Fig. 3. Low-temperature resistivity of a sample with x(Ba) = 0.75, recorded for different current densities

BaPb<sub>1-x</sub>Bi<sub>x</sub>O<sub>3</sub> polycrystalline thin films [5, 18]. Upon cooling from room temperature, the latter exhibit a nearly linear metallic decrease of  $\rho(T)$ , then a logarithmic type of increase, before undergoing the transition to superconductivity. One could, of course, speculate that in our samples a metal-to-metal structural phase transition occurs in one of the phases. The shift in the drop in  $\rho(T)$  with increasing current density (Fig. 3), however, would be hard to explain with such an assumption, while it supports our interpretation that we observe the onset of superconductivity of percolative nature, as discussed below. In BaPb<sub>1-x</sub>Bi<sub>x</sub>O<sub>3</sub>, the onset of superconductivity has been taken at the resistivity peak [18]. This assumption appears to be valid in percolative systems, i.e., in the thin films [18] consisting of polycrystals with grain boundaries, or when different crystalline phases with interpenetrating grains are present, as found in the Li-Ti-O [5] or in our Ba-La-Cu-O system. The onset can also be due to fluctuations in the superconducting wave functions. We assume one of the Ba-La-Cu-O phases exhibits this behaviour. Therefore, under the above premises, the peak in  $\rho(T)$ at 35 K, observed for an x(Ba) = 0.75 (Fig. 1), has

to be identified as the start to superconductive cooperative phenomena in the isolated grains. It should be noted that in granular Al, Cooper pairs in coupled grains have been shown to exist already at a point where  $\rho(T)$  upon cooling has decreased by only 20% of its highest value. This has been proven qualitatively [19] and more recently also quantitatively [20] by the negative frequency shift occurring in a microwave cavity. In 100 Å films, a shoulder in the frequency shift owing to 2D fluctuations was observed above the  $T_c$  of the grains. In our Ba-La-Cu-O system, a series of layer-like phases with considerable variety in compositions are known to exist [16, 21], and therefore 2D correlations can be present.

The granularity of our system can be justified from the structural information, and more quantitatively from the normal conductivity behaviour. From the former, we know that more than one phase is present and the question arises how large are the grains. This can be inferred from the logarithmic fingerprint in resistivity. Such logarithmic increases are usually associated with beginning of localization. A most recent example is the Anderson transition in granular Sn films [22]. Common for the granular Sn and our samples is also the resistivity at 300 K, lying in the range of 0.06 to 0.02  $\Omega$ cm, which is near the microscopic critical resistivity of  $\rho_c = 10 L_0 \hbar/e^2$ for localization. From the latter formula, an interatomic distance  $L_0$  in the range of 100 Å is computed, thus a size of superconducting grains of this order of magnitude must be present. Upon cooling below T<sub>c</sub>, Josephson junctions between the grains phaselock progressively [23] and the bulk resistivity gradually drops to zero by three orders of magnitude, for sample 2 (Fig. 1). At larger current densities, the weaker Josephson junctions switch to normal resistivity, resulting in a temperature shift of the drop, as shown in Fig. 3. The plateau in resistivity occurring below the 80% drop (Fig. 1) for the higher current density of 0.5 A/cm<sup>2</sup>, and Fig. 2 curve (6) may be ascribed to switching of junctions to the normal state.

The way the samples have been prepared seems to be of crucial importance: Michel et al. [21] obtained a single-phase perovskite by mixing the oxides of La and Cu and BaCO3 in an appropriate ratio and subsequent annealing at 1,000 °C in air. We also applied this annealing condition to one of our samples, obtained by the decomposition of the corresponding oxalates, and found no superconductivity. Thus, the preparation from the oxalates and annealing below 950 °C are necessary to obtain a non-perovskite-type phase with a limited temperature range of stability exhibiting this new behaviour. The formation of this phase at comparatively low temperatures is favoured by the intimate mixture of the compo-

nents and the high reactivity of the oxalates owing to the evolution of large amounts of H<sub>2</sub>O and CO<sub>2</sub> during decomposition.

#### IV. Conclusion

In the concentration range investigated, compounds of the Ba-La-Cu-O system are metallic at high temperatures, and exhibit a tendency towards localization upon cooling. Samples annealed near 900 °C under reducing conditions show features associated with an onset of granular superconductivity near 30 K. The system consists of three phases, one of them having a metallic perovskite-type layer-like structure. The characterization of the new, apparently superconducting, phase is in progress. An identification of that phase may allow growing of single crystals for studying the Meissner effect, and collecting specific-heat data to prove the presence of high  $T_c$ bulk superconductivity.

The authors would like to thank H.E. Weibel for his help in getting familiar with the conductivity measurement system, E. Courtens and H. Thomas for discussions and a critical reading of the manu-

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J.G. Bednorz K.A. Müller IBM Zürich Research Laboratory Säumerstrasse 4 CH-8803 Rüschlikon Switzerland

#### Note Added in Proof

Chemical analysis of the bulk composition of our samples revealed a deviation from the ideal LarBa ratios of 4 and 5.66. The actual ratios are 16 and 18, respectively. This is in agreement with an identification of the third phase as CuO. CCTTURENT Exh. #C

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VII

# university note book



NAME W. Kateley + C.C. Tsuei

SUBJECT\_

Low temp. T.C.

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VERNON MCMILLAN, Inc., ELIZABETH, N.J. 07208

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#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J.G. BEDNORZ ET AL. : Date: March 29, 1988

Filed: 05/22/87 : Serial No.: 06/053,307

Group Art Unit: 115 : Examiner: Dennis Albrecht

FOR: NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE,

AND METHODS FOR THEIR USE AND PREPARATION

# DECLARATION OF RICHARD L. GREENE WITH RESPECT TO HIGH TO SUPERCONDUCTIVITY

Commissioner of Patents and Trademarks Washington, D.C. 20231

Sir:

- I, Richard L. Greene, hereby declare and say that:
- 1. I received a Ph.D in physics from Stanford University in 1967, and joined the San Jose, California laboratory of the Research Division of International Business Machines Corp. in 1970. I was the Manager of a group conducting research on organic superconductors and have worked in the field of superconductivity for 20 years. I transferred to IBM Corp. research laboratory in Yorktown, New York, in July 1986, and continued thereafter to conduct research on superconductive materials. From about October 1986 to the present I have worked on high T<sub>C</sub> superconducting oxides.
- 2. At approximately the end of September first week of October, 1986, my manager, Chang C. Tsuei, showed me a copy of an activity report from the Zurich Research Laboroatory of IBM Corporation. This activity report described the work of J.G. Bednorz and K.A. Mueller and their discovery of new superconducting compositions. These materials were mixed copper oxide ceramics that exhibited an onset of superconductivity at a temperature significantly higher than the transition temperatures reported for previously known superconductors. Materials of this general class are now known in the art as high T

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superconductors. A true copy of this activity report is attached hereto and labeled Exhibit A.

- 3. Soon after reading this activity report and discussing it with Chang C. Tsuei, I called K.A. Mueller in Zurich and requested samples from him so that I could make measurements on these samples in the United States. This telephone call occurred approximately October 1 October 6, 1986. My intent was to begin a research project on these materials, as I was very interested in them based on my previous work in superconductivity. My plan at that time was to do experiments which would be complementary to those being conducted by Bednorz and Mueller in Zurich, so that a maximum amount of information could be obtained about these new superconducting materials. Based on the data in this activity report and on the results of susceptibility measurements described to me by Alex Mueller in the aforementioned telephone call, I believed that a new class of superconducting materials with T<sub>C</sub> greater than 30K had been discovered.
- 4. In approximately mid-October, 1986, Praveen Chaudhari, Vice-President, Science, at IBM's Watson Research Laboratory visited the Zurich IBM Lab. Based on my request for samples of the new superconducting material, Chaudhari told me that he had obtained them from Bednorz and Mueller and brought them back to the United States with him. These were about six samples in the Ba-La-Cu-O system. Chaudhari returned to the United States on or about October 20, 1986 and delivered these samples to me. Of these approximately six samples, they varied in the different amounts of La and Ba that were present. Only two of the samples were reported as being single phase materials.
- 5. Immediately upon receiving these samples, I was in contact with Bednorz and Mueller, via telephone and computer system links, in order to discuss with Bednorz and Mueller the experiments that I would conduct and also to obtain information from Bednorz and Mueller relative to the characteristics of the samples. I had planned to do specific heat measurements of the samples and also resisitivity versus temperature measurements in the presence of a magnetic field. Because of the importance that I attributed to this work, I worked substantially full time on these superconductor materials in order to further characterize them. My first specific heat measurements occurred approximately October 29 and 30, 1986, while I measured resistivity versus temperature in the Y0987-074

presence of a magnetic field in late November, 1986. Continuously throughout the period, October 20, 1986 - February, 1987, I worked on a daily basis to further characterize these materials. At all times, I was in contact with Bednorz and Mueller, exchanged data with them, and worked in close cooperation with them. They provided information to me about the characteristics of the material, as well as providing me up-to-date information concerning the data they had obtained about these materials. A true copy of my computer log from October, 1986 - January 12, 1987 is attached hereto and labeled Exhibit B. Excerpts which do not relate to superconductivity have been deleted. In this exhibit, the identifier for K.A. Mueller is "KAM", while the identifier for J. G. Bednorz is "BED". Bednorz and Mueller are located in Zurich, Switzerland and the computer node for them is ZURLVM1. My identifier is "RGREENE". This computer log details my ongoing computer dialogue with Bednorz and Mueller relative to theirs and my activities on the high  $\mathbf{T}_{\mathbf{C}}$  superconductor materials. addition to this computer correspondence, I also talked with Bednorz and Mueller via telephone.

- 6. During my specific heat measurements of these materials, as well as the measurements of resisitivity versus temperature in the presence of a magnetic field, I was assisted by Albert M. Torressen, who was a laboratory specialist. I also discussed my laboratory experiments with Chang C. Tsuei, S. von Molnar, Merril W. Shafer, Sung Il Park, Thomas Penney, and Arthur R. Williams.
- 7. The specific details of the apparatus and the data obtained in the specific heat measurements will be described in a separate statement by Albert M. Torressen, the laboratory specialist who worked with me to provide these measurements. Essentially, the specific heat of the apparatus was calculated to provide calibration and background specific heat, after which the sample was introduced into the apparatus and the total specific heat again measured. By subtracting the background specific heat, the specific heat of the superconducting sample is determined. This was done over a temperature range of approximately 2-50K.
- 8. The specific heat measurements of these superconducting samples were begun approximately October 21, 1986, and were conducted on a daily basis by me and Al Torressen through November and December, 1986.

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These specific heat measurements and the curves which were plotted are representative of these superconducting materials, and are also representative of the specific heat versus temperature plots obtained on present samples of superconducting high T<sub>c</sub> oxides.

- 9. In addition to the specific heat measurements described hereinabove and in the accompanying statement of Albert M. Torressen, I also performed measurements of resistivity versus temperature in the presence of a magnetic field, for the samples of Ba-La-Cu-O obtained from Bednorz and Mueller. The specific heat measurements were performed first on these samples, after which I measured resistivity versus temperature in an applied magentic field, in order to further characterize these samples. These resistivity measurements were done at the end of November, 1986, and the beginning of December, 1986. Exhibit C is a true copy of nine pages of my data notebook, together with a copy of the cover of this notebook entitled "Zurich oxide BLCO DATA (T,H)." The date "11/15/86" is also on the cover. Exhibit D is comprised of several pages of plots of resistivity versus temperature for these superconductor samples, as well as resistivity as a function of magnetic field at particular temperatures. In some instances, the RuO, sample holder is taken into account into the plots. Generally, these plots represent the graphical expressions of the data contained in Exhibit C. Exhibit E is a composite plot incorporating the different plots found in Exhibit C, and shows resistivity versus temperature for different values of applied magnetic field. I used this composite plot at a seminar that I gave to other researchers at the Yorktown lab on December 12, 1986.
- 10. In order to obtain the data listed in Exhibit C, I used a laboratory belonging to Stephan von Molnar. Albert M. Torressen, who reported to von Molnar, showed me the necessary equipment to make these measurements, and I preceded to make them on my own. However, many people were aware of these resistivity measurements and viewed the data, including both Thomas Penney and Albert Torressen. In addition, Thomas Penney observed me making these measurements and understood the procedure and nature of my laboratory work.
- 11. I have numbered the data pages of Exhibit C in red in the upper right hand corner. Page 1 describes the sample set-up that I used for these measurements and the background data in order to ready the

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apparatus for the resistivity versus temperature measurements. This sample was the BLCO -21 II, standing for Ba-La-Cu-Oxide material. Page 2 shows two views of the experimental apparatus and the calibration measurements made between particular terminals. The wires A, B, C and F are those which are also shown on page 1.

- 12. On pages 3, 4, and 5 I had listed the data that applied to the  ${\rm RuO}_2$  sample holder and the four point probe. The sample was contacted with indium contacts and copper wires were attached to the indium contacts for the measurements. Both DC and AC measurements were made. The resistance of the sample is  ${\rm R}_{\rm FC}$  which was measured at various temperatures with the applied magnetic field H equal to zero (page 4). Pages 5 9 show further measurements that were made at different temperature settings and applied magnetic fields. All of the data on these pages were taken by me and entered by me in this notebook.
- 13. The plot of resistance versus temperature in exhibit D is a plot for the data which was obtained December 3 December 5, 1986. Referring to Exhibit D, this plot shows the superconducting transition that begins to occur about 35K, where the transition shifts to the left in the presence of a magnetic field. This is an indication of a superconductor.
- 14. All acts performed by me as described hereinabove occurred in the United States.
- 15. I further declare that all statements made hereinabove are of my own knowledge and are true and that all statements made on information and belief are believed by me to be true. Further, I declare that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of a Patent Application or any patent issuing thereon.

Richard J. Greene

DATE: 30 MARCH 1988

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Greene

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Zurich Research Laboratory

8803 Rüschlikon, Switzerland

Teleprinter: ITPS CODE ZRL

IBM

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ACTIVITY REPORT MAY-JUNE, 1986

August 15, 1986

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MATERIAL SCIENCE

T. Schneider, Mgr.

SURFACE & MATERIAL SCIENCES E. Courtens, Mgr., Project 4181

**Novel Research** 

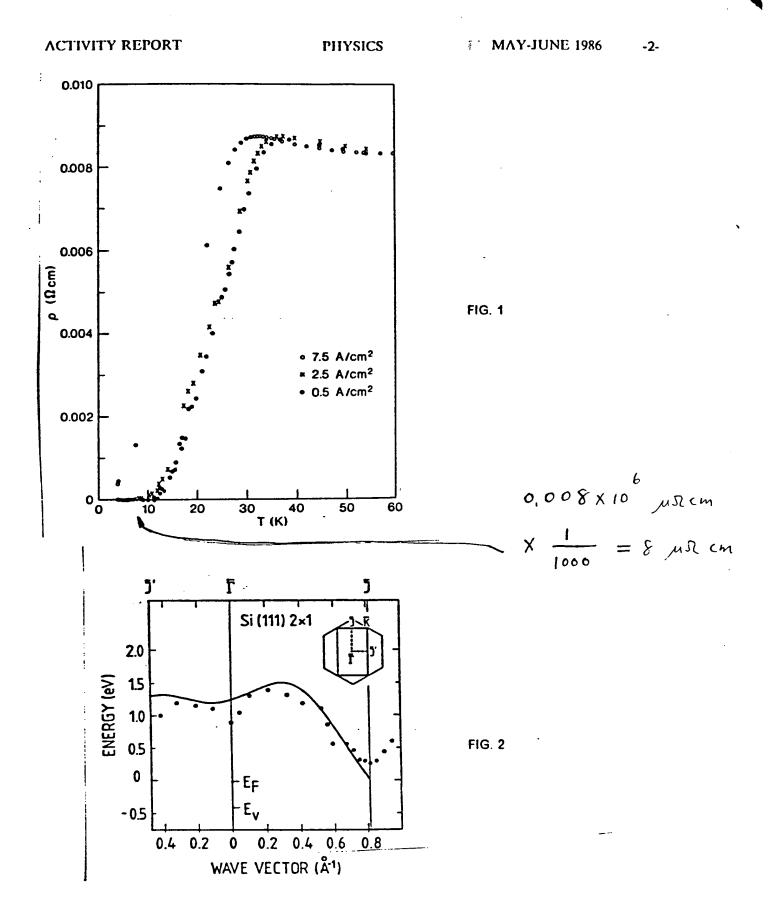
Possible High-T, Superconductivity in the Ba-La-Cu-O System

J.G. Bednorz and K.A. Müller (Project 4196)

We observed a steep decrease of resistivity in sintered Ba-La-Cu-oxide samples, with the highest temperature of the onset in the 35 K range (Fig. 1).

The Ba-La-Cu-O system exhibits a number of oxygen deficient phases with perovskite-like layer-type structures. These are characterized by mixed-valent copper ions (Cu<sup>2+</sup> and Cu<sup>3+</sup>) and itinerant electronic states. In addition one expects polaron formation induced by the strong Jahn-Teller effect of Cu<sup>2+</sup> in an octahedral oxygen environment. Thus our Ba-La-Cu-O system was anticipated to have considerable electron-phonon coupling and metallic conductivity.

Compounds with the composition Ba(x)La(5-x)Cu(5)O(5[3-y]) have been prepared in polycrystalline form. Samples with x < 0.2 and y > 0, annealed below  $900^{\circ}C$  under reducing conditions, consist of three phases, one of them a perovskite-like mixed-valent copper compound with  $K_2NiF_4$  type structure. Upon cooling, the samples show a linear decrease in resistivity, then an approximately logarithmic increase, interpreted as a beginning of localization. Finally a steep decrease by up to three orders of magnitude occurs, reminiscent of the onset of percolative superconductivity. The highest onset temperature is observed in the 35 K range. It is markedly reduced by high current densities (Fig. 1). The slow sensitivity decay towards low temperatures might possibly result from 2D superconducting fluctuations of perovskite layers of one of the phases present.



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Rick græne files Exh, B græne Date: 6 October 1986, 15:37:18 EDT From: RGREENE at YKTVMZ To: KAM at ZURLVM1

Alex:

Have you made any decision on my proposed specific heat experiment? I am anxious to try it. I think I can do it rather quickly after getting some samples. It may be difficult to see the transition near 30K because of a large phonon background but at the very least we could get a good estimate of the electron density of states and the Debye phonon contribution. Once I have a good specific heat between 2K and 40K I can make a better effort to resolve the electronic effect at Tc if it is small....I think I can see a 1% effect if the transition is not too smeared in temperature. Perhaps you would like to come to Yorktown and work with me on this experiment? Let me know. Best regards.

Rick

P.S. This is a good time for me to do some experiments on your exciting new compound since I am not heavily involved in other projects yet. I could get access to tunneling and neutron scattering equipment which would be very useful for seeing which phonons (if any) are involved in causing such a high Tc.

Date: 14 October 1986, 10:42:19 EDT From: RGREENE at YKTVMZ To: KAM at ZURLVM1

#### Hi Alex:

This note is to give you my user id and node on the VM system. You can see them above. When you send the samples for the specific heat experiment let me know via VM. My office at Yorktowm is 02-026. I will make the specific heat my highest priority and should be able to start the experiment as soon as the samples arrive. I will keep you informed on the progress of the experiment....it will probably take a few weeks to get reliable data assuming there are no unexpected problems.

As I said on the telephone you can ignore the sample dimensions sent to you by my manager C. Tsuei. He did not talk to me before sending you his VM note and he did not understand the requirements of the specific heat apparatus. What I need are samples to cover an area of 2mm x 2mm on the bolometer. Some extra samples will also be necessary in case we break or lose the primary samples. It would be best to not compress the samples or bind them together with any foreign materials which could alter the specific heat.

Best Regards,

Rick

Date: 15 October 1986, 15:02:34 EDT From: RGREENE at YKTVMZ To: SANDRO

Sandro; I have a court at 5pm. Do you want to play?

Rick

Date: 16 October 1986, 09:06:42 EDT From: RGREENE at YKTVMZ To: GGRIN

I'm thinking about taking my son to Mohansic tomorrow afternoon around 1:30. If you want to join us let me know.

Rick

Date: 16 October 1986, 13:27:32 EDT From: RGREENE at YKTVMZ To: JBMART

The reference is PRL 57, 1177(1986). I think a pressure experiment might be interesting. Let me call Chaikin and Brian first then I'll get back to you.

Rick

Greene

Date: 20 October 1986, 08:51:36 EDT From: RGREENE at YKTVMZ To: LOUGHRAN at ALMVMC

Hi Diane;

Starting to get cold around here but at least the sun is shining. Hope all is well with you. You can discard the Chaikin-Greene manuscript. It's been published and I have the reprints. Thanks for forwarding any remaining mail that comes to Almaden....it takes a long time for scientific types to know when one has moved. I'll be seeing you in Jan. Regards,

Rick

Date: 20 October 1986, 15:18:13 EDT From: RGREENE at YKTVMZ To: KAM at ZURLVM1

Alex;

The samples have arrived. They are bigger than I expected and all appear to be compressed pellets. Before I start on the specific heat I need to know a few things.

- 1. What is the difference between the samples marked I(red) and II(black)?
- 2. Have the samples been compressed with any foreign material, such as a binder?
- 3.Can the samples be cut without falling apart? If so do you recommend using a string saw or a razor blade or something else? Will water damage them?

4. Has the magnetic susceptibility been measured on any of these samples or on other samples from the same batch? I may want to measure the resistivity or susceptibility on these particular samples to make sure they exhibit the behavior you found before spending a big effort on the specific heat.

We are measuring the background specific heat of our apparatus up to 40K tomorrow...hopefully by the end of the week we will begin your samples so please call or send me via VM the answers to the above questions as soon as possible.

Best regards,

Rick

# Rick.

Date: 23 October, 1986, 13:43:17 EDT From: RGREENE at YKTVMZ To: BED at ZURLVM1 cc: KAM at ZURLVM1

Hi George , Alex;

Just a note to keep you informed of our progress. We are almost finished with the background specific heat. Tomorrow we will mount a 25mg slice of your sample BLCO2....it should take about a week to get the data in the earth's magnetic field. It probably will be necessary to also measure the specific heat in a magnetic field to accurately determine the superconducting contribution. Do you have any data on the critical field for this sample...if not we can measure it ourselves. Also I need to know if the samples you sent me are each a single phase....from your x-ray studies. I haven't received your preprint yet....perhaps some of my questions are answered there.

I will be away from the lab tomorrow and look forward to your response on Monday. Best regards.

Rick

Date: 23 October 1986, 14:00:11 EDT From: RGREENE at YKTVNZ To: MALOZEM

Alex:

Sorry I haven't gotten back to you but I have been very busy with two exciting experiments.....the specific heat of the new Zurich high

temperature superconductor and the 2D melting X-ray experiment with Paul which finally looks like it will work. I'm not really sure if there are any easily defined X-ray experiments that can be done to prove or disprove the nice model you presented this morning but I will think more about this. However given my present experimental committments it will be a few months before I could realistically do anything. Keep me informed. Thanks.

Rick

Date: 23 October 1986, 14:27:22 EDT From: RGREENE at YKTVMZ To: BED at ZURLVM1

George; Thanks for the Susceptibility info. I'm glad that I chose BLCO2 I for the first experiment. How wide in temperature is the para-diamagnetic transition in this sample? Regards.

#### Rick

Date: 23 October 1986, 18:58:35 SET From: j.g.bednorz
BED at ZURLVM1 To: RGREENE at YKTVMZ

Hi Rick

Here are the results of our susceptibility measurements, done on the samples You got from me. I'll give You the temperatures where the para- to diamagnetic transition occurs.

BLC02 I 32K BLC02 II 26-27K BLC08 I 13-14K BLC08 II 25-26K BLC021I 25K BLC021II 27-28K

So You don't need to involve somebody else with these measurements, which I prefere doing myself here. While typing this, I got the message that You send a note.

Salu George.

Date: 23 October 1986, 16:00:52 EDT From: RGREENE at YKTVMZ To: TFHEINZ

Tony;

I can't serve on the colloquim committee this year. I'll try to think of possible speakers however. Sorry and thanks for thinking of me.

Rick

Date: 23 October 1986, 17:32:09 EDT From: RGREENE at YKTVMZ To: GRANT at ALMVMC

Hi Grant:

Where have you been hiding? I need to talk to you since you didn't answer my last note. I'll be here on Monday..try me then.

WINDSING TO

VERMIJ

at Yorktown. Thanks and regards.

Rick

Date: 27 October 1986, 08:32:34 EST From: RGREENE at YKTVMZ To: GGRIN

Got your note too late....sorry I missed the big game. Let's try the

Date: 28 October 1986, 17:30:52 EST From: RGREENE at YKTVMZ To: BED at ZURLVM1, KAM at ZURLVM1

Hi George, Alex:

Did you get the last two notes that I sent you? I'm measuring BLC02 this week...nothing definitive yet. I'll keep you informed. What is the critical field of this sample? Is BLC02-I all the same phase? I haven't received your preprint yet...have you sent it? Best regards.

Rick

Date: 29 October 1986, 17:20:53 EST From: RGREENE at YKTVMZ To: GGRIN

Hey Rod;

Date: 29 October 1986, 16:41:22 SET From: j.g.bednorz
BED at ZURLVN1 To: RGREENE at YKTVMZ

Hi Rick,

Sorry for letting you wait so long to get an answer. Alex told me that he sent the reprint already 10 days ago. I have sent you a second one today, in case the letter got lost somewhere.

Concerning your questions:

From our measurements we can tell you that the critical field Hc2 is higher than 1.5 kG.

Now about the phases present in our samples:

BLC02 I 3 phases: °cub. perovskite/tetrag. perovskite/CuO
BLC02 II 2 phases: "/"/--BLC08 I 2 phases: "/"/--BLC08 II 1 phase: "/--BLC021I 2 phases: "/--BLC021II 1 phase: "/---

Best regards also from Alex

George.

Date: 30 October 1986, 09:20:04 EST From: RGREENE at YKTVMZ To: GRANT at ALMVMC

Hi Grantie;

I'm here ..where are you? Don't even have a phone answer any more. How come you didn't answer the questions in my last note.

As for the 3M meeting I am supossed to share a room with Torrance starting Sunday nite. I'm not sure if he's still coming or how long he's staying. Check with him and you can share the room with us or replace him. Let me know.

Greene

Date: 30 October 1986, 09:31:20 EST From: RGREENE at YKTVMZ To: BED at ZURLVM1, KAM at ZURLVM1

Hi Alex and George:

I just tried to reach you by telephone without success so here is a note. We have measured the specific heat(C) of BLCO2-I from 2-40K...the analysis is not yet complete but the prliminary data does not show any bump in C near or below 32K. However at this stage we could only see a bump or jump if it was greater than 10% of C so more accurate ex-

periments will be required. Since BLCO2-I is a 3 phase sample it was not a good choice for the measurement since I will not be able to analyze the data for density of states and Debye Temp. Do you know how much of each phase is present in this sample? Also is the cubic perovskite a metal or insulator?

It would be better if you had some single phase single crystals of the tetragonal phase. Is this possible? We could measure samples as small as a few milligrams.

Without crystals I am planning to measure BLCO8-II next since this is a single phase. Once you send me the info on the chemical composition and structure of this phase I can analyze the data and hopefully get results that we can publish. The measurements will take another week if all goes well. If we have to put on a magnetic field this will take several more weeks... specific heat data is tedious to obtain and analyze even with a computer.

Please answer the above questions as soon as possible. I am still waiting for your preprint...the first one must have gotten lost. Did you send it by external mail? Best regards.

# Rick

Date: 30 October 1986, 18:30:43 SET From: j.g.bednorz BED at ZURLVM1 To: RGREENE at YKTVMZ

Hi Rick,

BLC021II or BLC08II would be good to try.

 $\,$  BLC021II shows a more pronounced resistivity drop, as compared to the

sample I. BLCO8II I could not check till now.

The composition is Ba0.15 La1.85Cu04-x and Ba0.10La1.90Cu04-x respec-

tively. The structure of La2CuO4 is a layered perovskite of K2NiF4 type.

The pure material is orthorhombically distorted. Exchanging La by Ba

is leading, as we belief, to the formation of a tetragonal unit cell.

Our powder diffraction pattern can be indexed with a bodycentered lattice and a=3.79A and c=13.21A for xBa around 0.1. For crystals with xBa=0.02 I also checked the lattice parameters by single crystal

precession experiments. But here we aready have the problem. These crystals have been obtained from powders with xBa=0.1, so we have to expect seggregation and it will take a while, to get the crystals with a composition where the resistivity drop is observed in the powders.

To your question about the cubic perovskite, it shows metallic conduc-

tivity as well.

I really hope, that you get the preprint very soon.

# Best regards George.

Date: 30 October 1986, 15:06:11 EST From: RGREENE at YKTVMZ To: BED at ZURLVM1

George; Thanks for your quick answer to my questions. I forgot to ask you if you know the relative weight % of the 3 phases in sample BLCO2-I. If so I may still be able to get some useful information from the data we have taken so far.

I also just realized that you could send me the preprint via VM assuming it was typed on line. Please see if your secretary can do this. Thanks and regards.

Rick

Date: 3 November 1986, 16:58:28 EST From: RGREENE at YKTVMZ To: JERRYT at ALMVMC

# Hi Jerry;

All is set for our room at the Hyatt starting Sunday nite the 16th. I'm not sure when I'll arrive but they have your name attached to the room also and it's guaranteed for late arrival. See you there. I saw some article recently about an organic ferromagnet....I think in JETP letters. Do you know about that work?

Rick

Date: 4 November 1986, 17:00:53 EST From: RGREENE at YKTVMZ To: SANDRO

I cannot play tomorrow...sorry.Next week.If I can change my schedule tomorrow I will call you in the morning.

Rick

Date: 11 November 1986, 10:04:02 EST From: RGREENE at YKTVMZ To: BED at ZURLVM1

# Hi George;

No I have not given up...in fact I just tried to reach you by phone. My terminal is not working since I just changed my office so It may take me a little longer to respond to messages.

At any rate I have finished the specific heat measurement from 4-35K in zero magnetic field. It will take a few days to finish the data analysis but there is no obvious bump in the specific heat indicating superconductivity. This is not really too surprising given the very broad transition you have found in resistivity and susceptibility.

I expect to get some useful information from the data anyway but for this I need the exact composition of BLCO21-II.Is it Ba.15La1.85CuO(4-.15)? Please send this as soon as possible by VNET....I will get back to you and ALex later with more info. Regards.

Date: 12 November 1986, 09:19:56 EST From: RGREENE at YKTVMZ To: GRANT at ALMVMC

# Greene

Date: 13 November 1986, 13:54:49 EST From: RGREENE at YKTVMZ To: MALOZEM

# Alex:

I'll be happy to talk about the prospects of using magnetic X-ray scattering for thin films and interfaces. It will only be a summary of what has been done and my thoughts on what else could be done. The rest of your proposed program looks great. It's a good idea to have such an internal meeting.

Rick

Date: 14 November 1986, 10:17:09 EST From: RGREENE at YKTVMZ To: BED at ZURLVM1, KAM at ZURLVM1

# Hi Alex and George;

I will be away from the lab until 24 Nov. so I thought I would let you know the present status of the specific heat (SH) experiment and my future plans.

So far we have measured BLCO21 from 3-35K. There is no evidence for a bump in SH anywhere....to a 5% accuracy. I have analyzed in detail the data between 3-10K. Here the SH is linear on a C/T vs T2 plot. The intercept gives a value for gamma of 5.9 mj/mole-K2. This is a rather large value compared to other metals and suggests that most of the BLCO21 sample is in the normal state. However to be sure of this we must measure the sample in a magnetic field large enough to suppress the superconductivity. This we will start while I am away. Also we must run a test sample such as copper or silicon to know the accuracy of our gamma determination. All this will take 2-3 more weeks. As you see it takes considerable effort to do a reliable specific heat measurement which makes it very important that we have well characterized, single phase samples. As we discussed yesterday George it may be better to do the SH experiment on a bunch of single crystals if you can prepare them. Five mg of material should be enough to get reliable data.

We will also measure the critical field up to 9T via resistivity. I want to do this first so I have some idea of the field necessary to get the normal state at 3K....our SH apparatus has a field of 5T maximum.

I'll talk with you when I return. I am still quite excited about these new materials and hope that we can continue to collaborate on various experiments even if the specific heat does not give evidence for bulk superconductivity. I should remind you that it took many years of work before the BaPbBi Oxides were shown to be bulk Superconductors.

Best regards,

Rick

Date: 25 November 1986, 09:43:48 EST From: RGREENE at YKTVMZ To: PARKIN at ALMVMC

Hi Stuart;

Thanks for your note. I haven't heard from Helmut but he is probably very busy starting his new job. Haven't heard about the Japan proceedings either....are you still interested in organic metals? What is happening with your work on thin films? I expect to be out to San Jose sometime in January and you can bring me up to date.

Until then I am very busy with X-ray scattering and some other experiments on new inorganic materials. Have a good holiday season.

Best regards

Rick

Date: 25 November 1986, 10:35:50 EST From: RGREENE at YKTVMZ To: BED at ZURLVM1, KAM at ZURLVM1

Hi Alex and George;

I have returned from my trip and will once again start work on your new superconductor. This week is the Thanksgiving holiday so not much will happen until next week. The specific heat apparatus is now modified to make measurements in a magnetic field....however we must first calibrate and check that it works with some known material.

Please tell me what is happening with your studies of time dependent effects. Is the sample BLCO21 still good....we have not yet measured the resistivity in a field as a function of T but we plan to along with the specific heat experiment.Perhaps you should send me some new single crystals for the next experiment.....I don't want to waste time on a bad sample.

I would like to send an abstract to the March APS meeting on the specific heat results. Is this agreeable with you? The abstracts are due the end of next week (Dec.5) so let me know soon. At this stage there is not much definitive to say but I can still write a general abstract about specific heat and I'm sure I will have definitive results by the time of the meeting.

# Best Regards,

#### Rick

Date: 26 November 1986, 09:56:43 EST From: RGREENE at YKTVMZ To: KAM at ZURLVM1

# Hi Alex;

Are you sending your susceptibility preprint to people outside of IBM? If so Ted Geballe at Stanford would like a copy...he saw your paper in Z.Physic and called me to see if I knew about your work.

I can send him a copy if you are agreeable. Please let me know about this and more importantly the answer to my note of yesterday.

Best regards,

#### DOLTO

Date: 1 December 1986, 17:31:01 EST From: RGREENE at YKTVMZ To: KAM at ZURLVM1, BED at ZURLVM1

# Hi Alex and George:

Here is a draft of the abstract that I would propose submitting to the APS March meeting. Please make any changes or comments and let me know today. I look at this as a way to publisize your work in the USA and to present whatever specific heat results are obtained by March.

# POSSIBLE HIGH To SUPERCONDUCTIVITY IN THE Ba-La-Cu-O SYSTEM

We report measurements on new oxide superconductors of the composition La(2-x)Ba(x)CuO(4-y) with x<<1 and y>0. Polycrystalline samples with x=.15 show a resistivity drop of three orders of magnitude and a transition from Pauli paramagnetism to diamagnetism with an onset temperature between 30-35K. (ref 1 and 2....your two papers). The transition is complete by 10K and magnetic field studies suggest superconductivity of a percolative or granular nature. Our specific heat experiments indicate a large electron density of states but no evidence of a sharp jump near Tc---consistent with the small Meissner signal observed (2% of complete flux expulsion) and the broad transition width. These measurements, along with X-ray and critical field results, will be analyzed for the possibility of high Tc superconductivity in these new oxide materials.

The authors would be the three of us and Steve VonMolnar (whose apparatus I am using)....possibly I would add Al Torresen (Steve's assistant) without whom the specific heat experiments could not have been done.

The abstract could perhaps be a bit longer but there may not be much space after the authors and references are included.

Regards,

Rick

Date: 2 December 1986, 09:17:12 EST From: RGREENE at YKTVMZ To: KAM at ZURLVM1

Alex:

I scheduled your seminar for 8 Jan at 3:30pm....this was the only time I could get a room. Please send me a title and short abstract so I can get it on the lab calender as soon as possible. Do you need a hotel reservation? Regards.

Rick

P.S. Plan on saving some time on 9 Jan. to discuss our specific heat data. If you would like to go out together for dinner on the 8th let me know.

Date: 4 December 1986, 10:48:02 EST From: RGREENE at YKTVMZ To: BED at ZURLVM1

Hi George;

I just tried to telephone you. I am measuring the resistance and critical field of sample BLCO-21. So far it reproduces the data in your Z. Physik paper...I don't see a bump except perhaps near 25K (but I need to take more points). The surprising thing is that a small field (1000 Oe) increases the Resistance to the value at 25K but at higher fields (up to 7Tesla) there is almost no more change in R. Tell you more when I have more data....so far it suggests that doing the specific heat in low field will be useful.

Would you please send me whatever info you have on the structure of the superconducting phase i.e. a picture and a powder X-ray that gives the Bragg peak positions.

What have you learned about the time changes in these samples? I would like some fresh single phase samples for our next specific heat experiment...to begin at the end of next week. If you have single crystals that would even be better.....but I realize this is a difficult problem.

kegaras,

Date: 5 December 1986, 10:56:34 EST From: RGREENE at YKTVMZ To: ORR

OK. What are you up to? Dropin and see me sometime in vonMolnars lab.

Rick

Date: 5 December 1986, 11:10:42 EST From: RGREENE at YKTVMZ To: KAM at ZURLVM1, BED at ZURLVM1

Hi Alex and George:

I'm getting some good critical field results now although I still don't totally trust my contacts. The resistance vs. temp. follows your data but there seems to be two superconducting regions (perhaps 2 phases).. one below 22K and the other below 33K. The critical fields are very different in these two temperature ranges. The good news is that I am getting a critical field vs temp curve between 20-30K and this will alllow me to estimate gamma to compare with the specific heat gamma. Incidently the critical field at 4K is greater than 7Tesla (as expected for a high Tc material) so we may eventually want to go to the MIT magnet lab to measure it better.

The specific heat exp. is progressing nicely and we will be finished with all our calibrations next week. What sample do you recommend that I use based on your recent work?

As soon as I have collected and plotted all the critical field data I will send you a figure along with the abstract to the March meeting.

Best regards,

Rick

P.S. Please send me whatever info you have on the structure of the SC phases.

I am giving an internal journal club seminar on your resistivity and susceptibility  $$\operatorname{\mathsf{papers}}$.$ 

Date: 8 December 1986, 09:26:48 EST From: RGREENE at YKTVMZ To: LOUGHRAN at ALMVMC

uahhà untrada....zee hon tu nan (mahne)

Rick

Date: 3 December 1986, 16:39:02 SET From: KAM at ZURLVN1 To: RGREENE at YKTVMZ

Rick, here is the title and abstract for my seminar :

'Superconducting and Structural Properties of the BaLaCuO System'

Resistivity and susceptibility measurements as well as x-ray powder analysis carried out at the Rueschlikon laboratory will be described. The electric and magnetic data indicate the existence of a percolative superconductor with onset above 30 K. The newest magnetisation measurements as a function of temperature and field proove the presence of a superconductive glass. The highest Tc sampels correlate with an orthorhombic-tetragonal structural phase transition.

please check for the english, thanks

Alex

Date: 9 December 1986, 10:29:48 EST From: RGREENE at YKTVMZ To: GGRIN

Let's stick to our Weds tennis....4;30 right?

Rick

Date: 9 December 1986, 10:31:06 EST From: RGREENE at YKTVMZ To: POMERAN

Mel:

I can't take the court on Thurs. so why don't we just put off our game until next week.

Date: 9 December 1986, 10:37:27 EST From: RGREENE at YKTVMZ To: BED at ZURLVM1, KAM at ZURLVM1

Hi George , Alex;

÷."

Thanks for your note George. I will send you the Critical field data today. It seems to reproduce your low field results and has the data up to 7Tesla....I could go to 9T but will do that later. I assume from your note that you think that BLCO21 is still a single phase...is that correct? I will use this sample for the specific heat in a magnetic field.

I am a little puzzled by the critical field data...it suggests that your susceptibility data was measuring the superconductivity that occurs below 20K and the superconductivity above 20K may not be a bulk effect. It's also a litte disturbing that I measured such a large linear term in the specific heat in earth field....the measurements at 5T should clarify this however.

Can you tell me the density of the SC phase? I need this to estimate gamma from the critical field slope. Also what is your estimate of the value of the resistivity just above Tc? I assume a single crystal would be at least 10 times lower. Also I would like to know your estimate of the Pauli susceptibility above Tc from your data....this will give another estimate of gamma.

Thanks for sending me your info and figures of the structure..I hope it arrives before next Tuesday.

Best regards,

Rick

Date: 9 December 1986, 11:32:57 EST From: RGREENE at YKTVMZ To: MALOZEM

I don't know Creuzet that well but he seems to have done some good work and seems to know what he's talking about. I'm not sure how independent, creative or hardworking he is. What would he be doing here? How closely working with an RSM? Who with?

15

Date: 9 December 1986, 14:34:51 EST From: RGREENE at YKTVMZ To: POMERAN

Mel;

OK for Monday. SEe you there unless you hear otherwise.

Rick

Date: 8 December 1986, 18:56:40 SET From: j.g.bednorz BED at ZURLVM1 To: RGREENE at YKTVMZ

Hi Rick Sorry that you had to wait for the answer since Thursday. I've been in Germany since Friday. In November I told you on the phone, that something happened to that sample BLCO21II which I measured again one month after the first resistivity run. The resistivity curve showed a peak at 34K and a shoulder occurring around 25K after a 60 percent drop. At that time I was also surprised about the magnetic field dependence in the low temperature part. The resistance was increased by fields between 0-0.4 Tesla but seemed to saturate at values above, whereas the field dependence of the peak at 34K was smaller. It would be good to compare our results, especially as you have the possibility to go to higher fields than 0.7 Tesla, which is the limit for our resistivity system. Unfortunately I do not see the occurrence of a new phase related to the appearence of that shoulder in the resistivity.

Concerning your internal seminar, I will send you an X-ray powder spectrum and the structure of La2CuO4, which I've drawn already, using the information given in a German article. You can even have the viewgraphs. We should discuss questins about the structure at the phone.

Best regards George.

Date: 9 December 1986, 18:47:08 SET From: j.g.bednorz
BED at ZURLVM1 To: RGREENE at YKTVMZ

Hi Rick,

Thank you for your quick answer. I just discussed with our Japanese guest Masaaki Takashige, who is involved in the susceptibility measurements. First of all you should not be worried about about the susceptibility data shown in the preprint, because the samples shown there are not single phases. You will see from the X-ray pattern that the amount of the foreign phase can be very large, greater than in BLC021 I. Single phase means, that in the X-ray diagrams we only can detect the La2Cu04:Ba. The small susceptibility could indicate that only parts of that phase is superconducting, for instance an intragranular network. That is the reason, why we think the density of La2Cu04 (from the X-ray data = 7 g/ccm) would not lead to a correct estimation in your case. The Pauli susceptibility

of sample BLCO21II, this sample is not shown in the paper, shows a field dependence close to Tc, this dependence is getting weaker with increasing temperature, and we expect it to vanish 10 or 20 de- grees higher, but in case of this sample it has not been confirmed. I'll give you values at 32-33K for the mass susceptibility:

0.3 Kgauss 1.28 E-7 ccm/g 5.0 " 1.35 E-6 ccm/g

10.0 " 1.61 E-6 ccm/g Especially for the low field value we have to be aware of a large error.

For the resistivity value: My measurement (second one, where I realized the magnetic field dependence) showed a peak value of 7.36 E-3 Ohm cm.

Concerning the results of the Japanese group: Do you know more about it? How did they measure the 40% Meissner effect, did they measure ac or dc? Is something known about the magnetic field they applied? I think they believe the metallic perovkite phase is responsible for the superconductivity, whereas we found that the single phase samples containing Da2Cu04:Ba in the powder diffraction pattern; show the highest susceptibilities. You will get she aspies of the results as soon as they are plotted.

Best regards
George. can be very large, larger than
in BLCO21.Talking about single phase samples

les,

Date: 10 December 1986, 10:48:47 EST From: RGREENE at YKTVMZ To: GRANT at ALMVMC

I haven't forgotten you....just busy as hell with this 30K superconductor and can't think about anything else. Happy Holidays Turkey.

Looks like I won't have time to ski...too much physics to do.

1e

Greene

Exhibits
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# RG, unter

# 12/1/86

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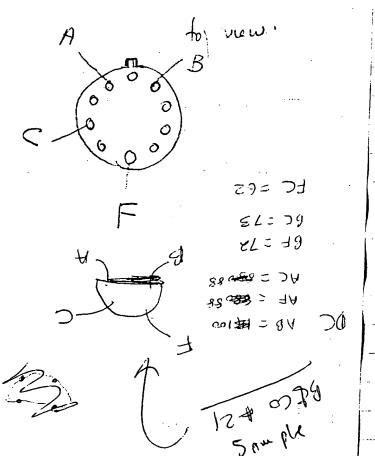
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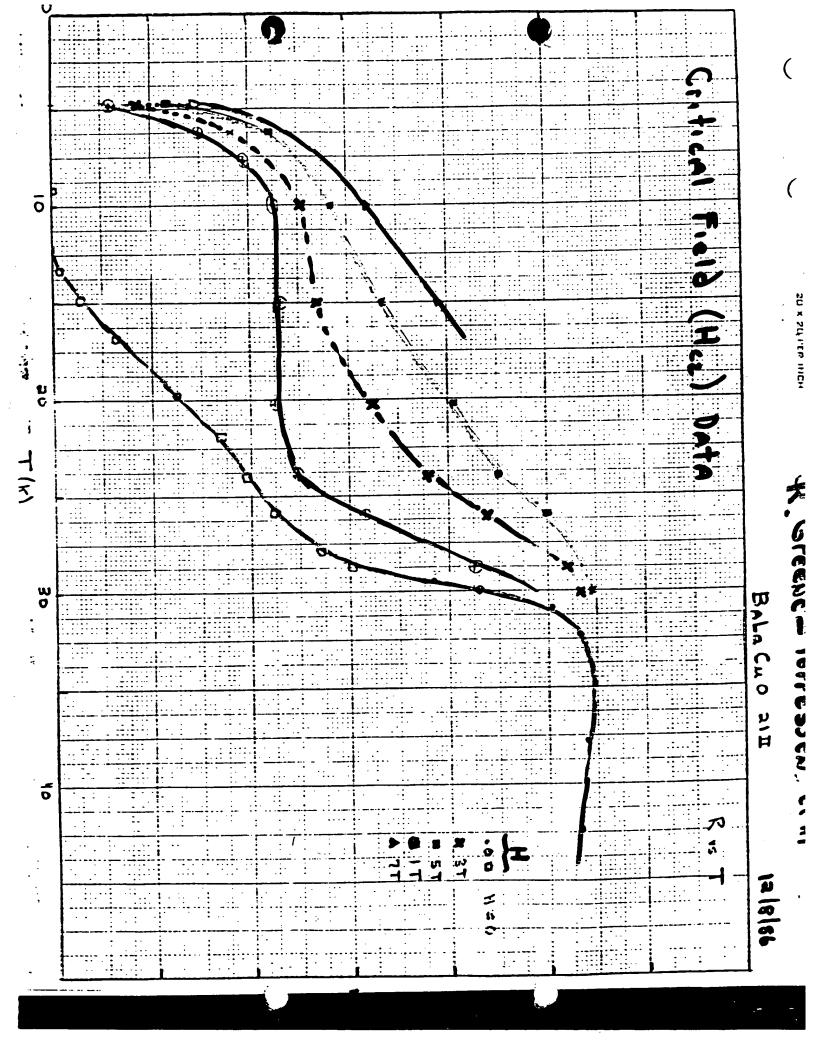
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THE OXYGEN DEFECT PEROVSKITE  $Bala_4^{Cu}_5^{O}_{13.4}$ , A METALLIC CONDUCTOR

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(Received March 14, 1985; Refereed)

#### ABSTRACT

A new oxygen defect perovskite BaLa<sub>4</sub>Cu<sub>5</sub>O<sub>13.4</sub>, characterized by a mixed valence of copper has been isolated; the parameters of the tetragonal cell are closely related to that of the cubic perovskite: a=8.644(4) Å =  $a_p$   $\sqrt{5}$  and c=3.867(3) Å =  $a_p$ . The X-ray diffraction study shows that the atoms are displaced from their ideal positions in the cubic cell, owing to the presence of ordered oxygen vacancies. The study of conductivity, magnetic susceptibility and thermoelectric power versus temperature shows that this oxide is a very good metallic conductor.

# INTRODUCTION

Oxygen defect perovskites, have been more extensively studied these last years owing to their potential applications in catalysis, electrocatalysis or as gauges (1-3). In this respect mixed valence copper oxides offer a wide field for investigation: several perovskites (4) or perovskite-related structures have been isolated (5-6). These materials in which copper takes several coordinations simultaneously and a valence state intermediate between II and III can intercalate large amounts of oxygen according to the oxygen pressure and the temperature. Their electron transport properties ranging from semi-conductive to metallic (7) are closely correlated to the amount of intercalated oxygen.

The present paper deals with a new oxygen defect perovskite  $BaLa_4Cu_5O_{13.4}$ , which is like  $La_3Ba_3Cu_6O_{14+\delta}$  (4) a mixed valence copper oxide but whose behavior is quite different.

# EXPERIMENTAL

### Synthesis

Samples were prepared in platinum crucible and in air from appropriate mixtures of dried oxides  $La_2O_3$ , CuO and carbonate  $BaCO_3$ . The mixtures were first heated a few hours at  $900^{\circ}\text{C}$ , ground and heated at  $1000^{\circ}\text{C}$  during several hours. They were then ground again, and mixed with an organic binder, compressed into bars and then slowly heated up to 1000°C. After 24 hours or more at 1000°C, the bars were finally quenched to room temperature, The use of a binder was necessary to avoid that the compressed bars break before

heating. In these conditions the compactness of bars was of about 80 %. Chemical analysis

In order to determine the oxidation state of the transition metal ions, chemical analysis were carried out by iodometric titration using KI and by reduction in a flow of 25 % hydrogen in argon up to about 1000°C using a SETARAM microbalance for weight loss measurements.

# Structural analysis

The cell parameters were determined from X-ray powder diffractogramms registered with a Philips goniometer using Cu  $\rm K_{\alpha}$  radiation. The space group was determined by electron diffraction using a JEOL 120CX electron microscope.

# Magnetic and electrical measurements

The magnetic susceptibility was measured on powders by the Faraday method in the range 80-300K using a Cahn RG microbalance.

The conductivity was measured by the four points method on sintered bars. It was calculated by measuring the intensity/voltage ratio between the points in each current circulation direction in order to minimise the dissymetry effect between the contacts. The Seebeck coefficient was measured on the same sintered bars hold between two Pt heads.

Measurements were carried out up to 600K under an helium pressure of 200 mbars for T < 290K and in air for T > 290K in order to avoid possible departure of oxygen.

# RESULTS AND DISCUSSION

The scanning of the system La<sub>2</sub>O<sub>3</sub>-BaO-CuO for the compositions corresponding to the molar ratio (La + Ba)/Cu = l allowed us to isolate a perovskite for La/Ba = 4. The X-ray diffraction pattern of this compounds presents besides the intense lines which can be indexed in a cubic perovskite cell, extra lines which are rather weak. This feature is confirmed by the electron diffraction study, which shows superstructure reflections, leading to a tetragonal cell whose parameters are related to the cubic perovskite subcell (a<sub>p</sub>) as follows:

 $a = a_p \sqrt{5}$   $c = a_p$ 

all the lines of the X-ray diffraction patterns can be then indexed with accuracy in the tetragonal system with a = 8.644(4) Å and c = 3.867(3) Å. No reflection conditions are observed. The analysis of the oxygen content leads to the formulation BaLa4Cu5013.4 involving the presence simultaneously of Cu(II) and Cu(III) in spite of the presence of numerous oxygen vacancies (10.7 %). The measure of the density by pycnometry in benzene at 25°C(dexp = 7.05) confirms this composition for one mole per cell (dca1c = 7.03). Thus it appears that the oxide BaLa4CuII2.2CuIII2.8013.4 1.6 exhibits a great similarity with the oxygen defect perovskite Ba3La3CuII5.28CuIII previously described. However, this compound is very different from Ba3La3Cu6014+& from the point of view of the oxygen intercalation: no intercalation or desintercalation of oxygen has been observed by annealing this phase at low temperature (400°C to 500°C) and under different oxygen pressures up to 1 bar contrary to Ba3La3Cu6014+&. In the same way, no oxygen loss has been observed by TGA measurements for temperatures up to 650, 750 and 850°C and under oxygen pressures of 0.02, 0.2 and 1 bar respectively.

Taking into account the fact that the fundamental lines are indexed in a cubic perovskite cell and are strong with respect to the superstructure lines it was interesting to determine whether the metallic atoms were displaced from their ideal positions in the perovskite, or if the superstructure lines

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were only due to the ordering of oxygen vacancies. However, owing to the small amount of oxygen vacancies it was not likely to determine the distribution of the oxygen atoms by X-ray powder diffraction. Thus the structural study was undertaken for the composition  $La_4BaCu_5O_{15}$  just to determine the positions of the atoms with respect to the cubic perovskite subcell. Eight space groups were possible, they were reduced to three P4,  $P\overline{4}$  and P4/m taking into account the analogy with the perovskite structure. Calculations were carried out in the most symmetrical space group P4/m. For a ranging from 0 to 48°, 37 peaks i.e. 84 hkl were registered. The disparity between Fhkl and F hkl led us to introduce 139 hkl in the calculations. In the same angle r nge 13 diffraction peaks (14 hkl) were indexed in the cubic perovskite cell with a = 3.867 Å, and used in a calculation with the atoms in the ideal positions of the cubic perovskite cell, involving only a refinement of the thermal factors B; this first refinement led to a discrepancy factor  $R = \Sigma | I \text{ obs.} - I \text{ calc} | / \Sigma I \text{ obs.}$  of 0.066 with B(La, Ba) = 1.2 Å<sup>2</sup>, B(Cu) = 2.6Å B(O) = 3.9 Å<sup>2</sup>. The high B values let us think that the atoms were displaced from their ideal positions. A calculation carried out with all the intensities in the P4/m space group and the same ideal positions and overall  $B = 1 \text{ Å}^2$ (Table la), led to R = 0.35 in agreement with this point of view. Starting from these ideal positions, and assuming a statistical distribution of the oxygen vacancies in the oxides BaLa<sub>4</sub>Cu<sub>5</sub>O<sub>13.4</sub>, the R factor was lowered to 0.083, by refinement of the atomic parameters, the B factor being fixe at 1  ${\rm \AA}^2$ . From the final atomic parameters (Table 1.b) it can be seen that several atoms are displaced from their ideal positions in the cubic perovskite.

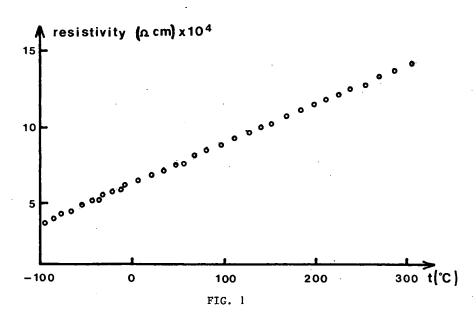
TABLE 1 Atomic Parameters of BaLa<sub>4</sub>Cu<sub>5</sub>O<sub>13.4</sub> (a) ideal positions (b) after refinement in the space group P4/m

Atom	Site	x	(a) Y	z	x	(b) Y	Z
Ba, La	1(d)	0.5	0.5	0.5	0.5	0.5	0.5
Ba, La	4(k)	0.1	0.3	0.5	0.124(1)	0.277(1)	0.5
Cu	l(a)	0.0	0.0	0.0	0.0	0.0	0.0
Cu	4(j)	0.4	0.2	0.0	0.415(3)	0.168(2)	0.0
0	1(Ъ)	0.0	0.0	0.5	0.0	0.100(2)	0.5
0	2(e)	0.0	0.5	0.0	0.0	0.5	0.0
0	4(j)	0.3	0.4	0.0	0.261(7)	0.384(8)	
0	4(j)	0.2	0.1	0.0	0.229(8)	0.063(6)	0.0
0	4(k)	0.4	0.2	0.5	0.428(10)	0.155(6)	0.0

Further refinements, concerning the ordered distribution of oxygen in this structure, which is most probable, were not carried out due to the rather low content of oxygen vacancies, and the too small number of refle-

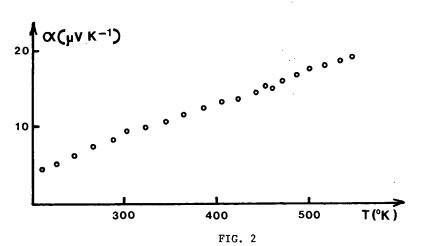
This oxide is a very good conductor: its conductivity is about 1.6  $10^3$  ( $\Omega$  cm)<sup>-1</sup> at room temperature. Figure 1 which represents the resistivity p versus temperature, shows that this oxide exhibits a metallic conductivity from 200 to 600K. The  $\gamma$  value deduced from the equation  $\rho=\rho_0(1+\gamma t)$  $(\gamma = 4.1 \ 10^{-3} \ c^{-1})$  is very close to that of free electrons  $(\gamma = 3.7 \ 10^{-3} \ c^{-1})$ .

The molar magnetic susceptibility is very weak and nearly independent of temperature. This suggests a Pauli paramagnetism which is characteris-



Resistivity plotted as a function of temperature

tic of delocalized carriers. The Pauli susceptibility (8) calculated with  $m^{\text{M}}/m$  = 1 and for one carrier per Cu(III) ( $\chi_{\text{M}}$  = 5.3  $10^{-5}$  e.m.u) is however one order of magnitude lower than the experimental value :  $\chi_{\text{M}}$  = 6  $10^{-4}$  e.m.u. The increasing of the Pauli susceptibility up to the experimental value needs  $m^{\text{M}}/m$  = 10. This suggests a strongly correlated carriers gaz (degenerated spin polaron gaz) which was introduced by Mott (9) to explain the magnetic susceptibility of LaCuO\_3 and LaNiO\_3 which are metals (10). At room temperature, the Seebeck coefficient is also very weak and positive ( $\alpha$  = 9  $\mu$ VK $^{-1}$ ) and increases slightly with temperature ( $\alpha_{500\text{K}}$  = 18  $\mu$ VK $^{-1}$ ) (Fig. 2). This



Evolution of the thermoelectric power as un function of absolute temperature.

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shows that the conduction mechanism is not thermally activated in this temperature range and that the behavior is metallic. The positive sign of the Seebeck coefficient indicates a hole conduction and that the conduction band is more than half filled.

A structural study by high resolution electron microscopy and by neutron diffraction will be carried out in order to determine the distribution of the oxygen vacancies in this oxide. This should allow an extensive interpretation of the electron transport properties of this compound.

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and B. RAVBAU, Mat. Res. Bull., 1975, 10, (Received November 30, 1983)

> Repue de Chimie minérale, t. 21, 1984, p. 407

## in mixed valence copper oxides related Oxygen intercalation to the perovskites

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Chimie et Physique des Solides, L. A. 251, ISMRa, Equipe Ozydes du Laboratoire de Cristallographie, Claude MICHEL and Bernard RAVEAU Université, 14032 Caen Oedex, France.

(A = Ca, Sr, Ba). These mixed valence copper oxides, characterized by the presence of series of compounds: Ba, La, Cu,O1+8. La,-,A1+,Cu,O4-x/3+4 and Las-xA,CuO4-x/8+8 cies in (001) planes of these structures makes that two of these families: Ba, Ca, Cu, O14+8 Sr0-perovskite: Sr,Ti,O, and K,NiF, respectively. The localization of the oxygen vacanrelated to that of the perovekite, and to those of the two members of the intergrowths Qu(II) and Qu(III) simulianously are oxygen defect compounds whose structure is closely on the oxygen pressure and on the nature of the oxides. conductive to a p type semi-metallic or metallic state is indeed observed which depends influenced by the intercalation process. A progressive transition from a p type semiand La-A1+xCueOe-x13+8 can be considered in their most reduced state as exides with The electrical properties of these phases are described and discussed: they are strongly low dimensionality. The influence of oxygen intercalation on the structure is described. Abstract. - Intercalation of oxygen in ternary copper oxides has been studied for three

pour trois séries de composés : Ba<sub>s</sub>La<sub>s</sub>Ou<sub>6</sub>O14+8, Las-xA1+xOu<sub>6</sub>O6-x/8+8 nature des oxydes, est en essel observée. métallique ou métallique de même type, qui dépend de la pression d'oxygène et de la calation. Une transition progressive d'un état semi-conducteur de type p à un état semiphases sont décrites et discutées : elles sont fortement influencées par le processus d'interl'Intercalation d'oxygène dans la structure est décrite. Les propriétés électriques de ces dans seur état le plus réduit, comme des oxydes de basse dimensionalité. L'influence de de ces familles : BagLagOugO11+8 et Lag-xA1+xOugO1-xfe+8 peuvent stre considérées, localisation des lacunes auloniques dans les plans (0 0 1) de ces structures fait que deux deux membres de la série d'intercroissances pérovakite-Sro : Sr, Tl, O, et K, Nif. La dont la skructure est étroitement liée respectivement à calle de la pérovikite et à celles des par la présence simultanée de Cu(II) et Cu(III), sont des composés déficitaires en oxygène Lag-xAxO10,-x1948(A = Ca, St, Ba). Ces oxydes de cuivre à valence mixte, caractérisés Résont. — L'intercalation d'exygène dens les exydes ternaires de cuivre a été étudiée ደ

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MICHEL AND B. PAVEAU

## INTRODUCTION

electron transport properties of these phases are discussed oxides related to the perovskite [11-13] and belonging to the systems La<sub>2</sub>O<sub>3</sub>. AO-CuO with A = Ca, Sr, Ba. The influence of oxygen intercalation on the and at low temperature (T  $\sim$  400-500° C) in three series of ternary copper deals with the soft intercalation of oxygen, i. e. at low pressure  $(p \leqslant 1 \text{ atm})$ formation of Cu(III) in normal pressure conditions [9-10]. The present paper compounds. However, the presence of A elements like barium favours the to several kbars (3-8] are most of the time necessary to synthesize these than those with Cu(11), since oxygen pressures ranging from 1 bar [4-7] coordination in similar structures as shown from previous works on especially considered owing to their possibility to take the same octahedral take several coordinations—octahedral, square pyramidal, square planar copper oxides are very good candidates, owing to the ability of copper to atoms which participate to the framework of the oxide. In this respect change of the oxidation state and of the coordination number of the metallic Ternary oxides AxCuyO3 containing Cu(III) are more difficult to prepare  ${
m La_2Cu^{II}O_4}$  [1-2] and  ${
m LaSrCu^{III}O_4}$  [3], which are isostructural with  ${
m K_2NiF_4}$ . and several oxidations stades: +1, +2, +3. Cu(II) and Cu(III) must be phase in their «oxidized» state. This phenomenon supposes a reversible able to absorb oxygen from atmosphere tending towards a stoichiometric exhibit rather large oxygen defects in their a reduced n form, and must be ties sensitive to the oxygen content. Thus it appears that such oxides musi tions such as electrocatalysis, or gauges for materials with electrical proper with  $O_2$  in air or in a gaseous atmosphere can be used for different applica-Intercalation of oxygen in an oxide, by a simple reversible exchange

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# STRUCTURAL CONSIDERATIONS

Three families with an oxygen defect structure have been isolated in the systems  $La_2O_3$ -AO-CuO:

- The oxygen defect perovskites La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14+5</sub>.
- The oxygen defect intergrowths Sr<sub>3</sub>Ti<sub>2</sub>O<sub>7</sub> type,

 $La_{2-x}A_{1+x}Cu_2O_{6-x/2+\delta}A = Ca$ , Sr

The oxygen defect intergrowth K2NiF4 type, La2-xA2CuO4-x/2+1.

The most reduced form which has been isolated for the defect perovskites La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14+1</sub>, corresponds to the formulation La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14+1</sub>. Its

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structure (fig. 1) can be described as an ordered oxygen defect perovskite. All the metallic sites corresponding to the stoichiometric perovskite are occupied by copper ions and lanthanum and barium ions respectively, whereas only 7/9 of the anionic sites are occupied in an ordered manner.

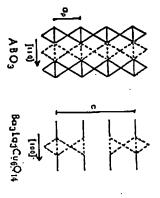


Fig. 1. — Schematic structure of a stoichiometric ABO, perovskite and the defect oxygen perovskite Ba<sub>2</sub>La,Cu<sub>2</sub>O<sub>11</sub>.

octahedra, parallel to (001) are preserved, that one apex out of two is ensured by lanthanum ions located at z=1/2. It is remarkable that such an considered as a true layer structure: double defect perovskite layers hedra are missing at z = 1/2. It results that this reduced form can be missing at the levels z = 1/6 and 5/6, whereas all the apices of these octa-Considering the tetragonal cell of this compound ( $a \simeq a_p \sqrt{2} = 5.525 \text{ Å}$ , corresponding to the formulation La, Ba, Cu, O14.10. The most reduced at 1 000° C for 24 h the mixture of La2O3, CuO and BaCO3 and quenching defect content. Site potential calculations confirm that the Cu3+ ions are oxide is characterized by a high Cu(III) content in spite of the high oxygen pyramids CuO, and square groups CuO, are observed whose cohesion is Ba<sub>1.5</sub>La<sub>6.5</sub>Cu<sub>3</sub>O<sub>7</sub> | 3 built up from corner-sharing, octahedra CuO<sub>6</sub> square phase La, Ba, Cu, O14.05 is then synthesized by annealing the sample the samples at room-temperature a slight excess of oxygen is indeed observed located preferentially on the octahedral sites. It must also be noted that this La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14.10</sub> at 400° C under low oxygen pressure ( $\sim 5.10^{-3}$ limit compound has not really been synthesized. By heating in air = 11.721 Å), it can indeed be seen that the basal planes of the

during several hours. The deviation from stoichiometry in the oxides  $\text{La}_{2-x}A_{1+x}Cu_2O_{6-x/2+\delta}$  is more complex owing to the possibility of substitution of calcium or strontium for lanthanum, in a small homogeneity range  $(0 \le x \le 0.14$  for

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forming NiF<sub>6</sub> octahedra (fig. 2 b). Like La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14+b</sub>, La<sub>2</sub>SrCu<sub>2</sub>O<sub>6</sub> is structure: the latter corresponds indeed to the superposition of two square pyramid layers. Such slabs are in fact derived from the K2NiF4 slabs are themselves an intergrowth of SrO-type layers and comer-sharing slabs | LaSrCu206 | parallel to (001) whose cohesion is ensured by ture with low dimensionality. It can indeed be described as built up from Nevertheless this oxide, like La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14</sub>, must be considered as a struc-Cu(II) exhibits here only one coordination which is square pyramidal. resulting configuration of the framework is different from La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14</sub>: z=1/2 all the apices of the oxygen octahedra are missing. However, the octahedra parallel to (001) are also preserved whereas at z=0 and similarity with those observed for La, Ba, Cu, O,4: the basal planes of the perovskite layers and SrO type layers. The perovskite layers exhibit some  $|K_2Ni_3P_6|_{\infty}$  slabs which would share the face of their square pyramids, thus, this oxide can be considered as an intergrowth of double oxygen six anionic sites out of seven are occupied by oxygen in an ordered manner; on the Ti<sup>4+</sup> sites, La<sup>3+</sup> and Sr<sup>2+</sup> ions are located on the Sr<sup>2+</sup> sites, whereas closely related to that of Sr<sub>3</sub>Ti<sub>2</sub>O<sub>7</sub> (fig. 2a). Cu<sup>2+</sup> ions are indeed located Its tetragonal cell (a = 3.865 A, c = 19.887 A), corresponds to a structure been isolated in this family corresponds to the formulation La2SrCu2O6. strontium and x = 0.10 for calcium). The most reduced oxide which has and La<sup>3+</sup> ions located at z = 0 and z = 1/2. The |LaSrCu<sub>2</sub>O<sub>6</sub>| $_{\infty}$ 

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Fig. 2.

a) Schematic structure of Sr<sub>8</sub>Ti<sub>2</sub>O, and La<sub>2</sub>SrCu<sub>2</sub>O, (projection on to (100) plane) showing the oxygen vacancies.

 Schematic representation of K<sub>2</sub>Ni<sub>2</sub>F<sub>4</sub> slabs sharing the square faces of the NIF<sub>4</sub> pyramids to give the K<sub>2</sub>NiF<sub>4</sub> structure.

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characterized by a great stability in spite of its oxygen defect structure: it is indeed synthesized by heating the stoichiometric mixture of CuO, La<sub>2</sub>O<sub>3</sub> and SrCO<sub>3</sub> at 1 050-1 100° for 24 h in air and by quenching them at room temperature in order to avoid their oxidation at lower temperature. Contrary to La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14</sub>, copper is in its lower oxidation state, Cu(II) in this oxide.

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The oxides  $\text{La}_{2-z}A_z\text{CuO}_{4-z/2+\delta}$  exhibit an oxygen defect  $\text{K}_2\text{NiF}_{\delta}$  type structure involving different coordinations of copper: octahedral, square pyramidal and eventually square planar (fig. 3). Their oxygen content

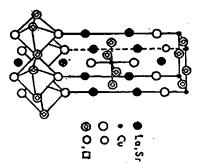


Fig. 3. — Perspective view of the structure of the oxides La<sub>3-x</sub>Sr<sub>x</sub>CuO<sub>4-x/p+8</sub> with oxygen vacancies located in the basel plane of the octahedra.

depends on the nature of the A ions (A = Ca, Sr, Ba) and on the substitution rate x which can lead to wide homogeneity ranges:  $0 \le x \le 0.20$  for A = Ca and Ba and  $0 \le x \le 4/3$  for A = Sr. The most reduced phase which exhibits the highest deviation from stoichiometry has been synthesized in the case of strontium for x = 4/3: La<sub>2/3</sub>Sr<sub>4/3</sub>CuO<sub>3.33</sub>. Contrary to the two other series, the oxygen vacancies are located in the basal plane of the coetahedra which are parallel to the (001) plane of the tetragonal cell (a = 3.759 Å, c = 12.907 Å). It must also be emphasized that this type of localization of the oxygen vacancies is always observed whatever the nature of the A ions, and whatever the rate of substitution x may be. However, symmetry changes and order-disorder phenomena in this plane may appear according to the nature of A and x value (table I). So, the calcium and barium oxides are characterized by a monoclinic distortion of the tetragonal  $K_2$ NiF4 structure, whatever the x value may be  $0 \le x \le 0.20$ ; the same

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corresponding to these homogeneity ranges exhibit an orthorhombic cell related to that of  $K_2NiF_4$  in the following way:  $a \simeq b \simeq aK_2NiF_4 \sqrt{2}$ is true for the strontium compounds with  $0 \le x \le 0.10$ . Thus, the oxides and c ≃ cK2NiF4.

The oxides La1-,A,CuO4-x12+3: crystallographic data and analytical results (quenched materials). I STRYI

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These a paran		3	1.20	1.0	0.00	9 6	0 66	0.50	رز. در	0.23	9.70	0.63	2	0.2	35			0.2	9.5		200	0	,	н	
eters are those	6.0	2 6	>	0.005	0.06	2.5	0.76	010	<u> </u>	0.07	0.04	9.5		0.05	0.02	10.0		0.0 0.0 0.0	0.02	9.01	2		•	<b>&gt;</b> -	
(*) These a parameters are those of the tetragonal subcell	3.759 (*)	V X 3.70	\$ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \	3.767 (*)	3.769	3.//3		3 776	3 774	3.775	3.774	1555		5.357	3.156	5.151		5.355	5.359	3.361	2.500	777.5	•		ة راق رق
al subcell.												5.168	0.000	4 380	3.J&S	5.387			5.364	5.380	3.402	3		•	Cell parameters (A)
	12.907	12.940	13.00%	13 000	13.070	13.168 6	13.210	10.404		11 347	13.231	13.200	13.410	3	13.174	13.150	15.520	13 170	IJ.245	LJ.201	13.149	:	ņ		<del>\</del>
	1,200	1,200	1,200	1 1 1 1	- 175	1.170	1,160	5.5	1,58	3	3	1.000	,,,,,,		1 100	1.18	1,100		3	1,700	, 100 1, 100		Separate of	temps-	Heating

and 2 of a series of oxygen defect intergrowths between perovskite and in that they can be considered as being respectively the members n = 1very closely related to the second series formulated La<sub>2-x</sub>A<sub>1+x</sub>Cu<sub>2</sub>O<sub>6-x/2+ $\delta$ </sub> belonging to the second series. The oxides La2-A2CuO4-22+, appear at 1 200° C and quenching the phase at room temperature. It appears here c remaining unchanged ( $c \simeq c K_2 NiF_4$ ). These oxides are very stable in is prepared by heating a mixture of the compounds La2O3, CuO and  $SrCO_3$ spite of the high deviation from stoichiometry; for instance La2/3Sr4.1 CuO, 13 with  $a = b \simeq naK_2 \text{NiF}_4$ , n ranging from 1 to 6 according to the composition, that the most reduced phase exhibits also only Cu(II) like La<sub>2</sub>SrCu<sub>2</sub>O<sub>6</sub> appear on the electron diffraction patterns which involve tetragonal cells  $(a \simeq aK_2NiF_4)$ ;  $c \simeq cK_2NiF_4)$ , whereas for  $1 \le x \le 4/3$ , superstructures metry similar to that of  $K_2NiP_4$  or LaSrCuO<sub>4</sub> [3] for 0.10 < x < 1On the other hand, the strontium compounds exhibit a tetragonal sym

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SrO structures, corresponding to the general formulation A,+1B,O3,++, an oxide with low dimensionality. two other series in that it cannot be considered in its most reduced form as However the behaviour of La2-A2CuO4-x/2+d is very different from the

## OXYGEN INTERCALATION AND DESINTERCALATION: INFLUENCE ON THE STRUCTURE

different oxygen pressures. annealing of the materials at low temperature, i. é. 400° C-500° C, under Oxygen can be intercalated in these three series of oxides by simple

absorb rather important oxygen amounts by annealing the samples at of La, Ba, Cu, O14.10 or from more oxidized compounds by simply annealing table II. In the same way, oxygen can be desintercalated from the structure 400° C under oxygen pressures ranging from 10-2 to 1 bar as shown from (table II). Thus it appears that the intercalated oxides La, Ba, Cu, O, 4+4 the samples always at 400° C under lower oxygen pressure, 5.10<sup>-3</sup> bar The oxygen defect perovskile La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14.10</sub> synthesized in air can

Evolution of  $\delta$  as a function of the oxygen pressure after annealing the oxide La<sub>3</sub>Ba<sub>2</sub>Cu<sub>6</sub>O<sub>14.10</sub> at 400° C. II STRVI

	٠,	), (bar)	
•	0.03	5.10-	
	0.19	10-1	
	0.25	2.10-	
	0.31	5.10-1	
	0.33	0.1	
	0.37	0.2	
	0.43	-	

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described above, forming corner-sharing ribbons of CuO, octahedra running between two square pyramids  $CuO_5$ , i. e. at z=1/2, between the layers tion; however site potential calculations [14], assuming that Cu3+ is octapossible to localize the additional oxygen in the structure by X ray diffrac sized oxide is observed when & tends towards zero. It is of course not c = 11.729 A, whereas no parameter change with respect to the air synthe meters very similar to those of La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14.10</sub>, a = 5.529 Å and most oxidized compound, La, Ba, Cu, O14.43, is characterized by paraof oxygen in this structure does not influence the cell parameters, since the exhibit a rather wide homogeneity range  $0.05 \leqslant \delta \leqslant 0.43$ . The intercalation be discussed further, are in agreement with this hypothesis. The fact that hedrally coordinated, show that this additional oxygen should be located along c. The electron transport properties of these compounds, which wil

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the c parameter does not vary, in spite of the intercalation of rather great amounts of oxygen is easily explained by the high oxygen defect content in the structure: the slabs | Ba<sub>1.5</sub>La<sub>0.5</sub>Cu<sub>3</sub>O<sub>7</sub>|<sub>10</sub> exhibit, themselves, oxygen defects, which may favour slight displacements of the copper and oxygen atoms along c during oxygen intercalation, between the slabs, without changing the c parameter.

The oxygen intercalation in the second series,  $\text{La}_{2-x}A_{1+x}\text{Cu}_2\text{O}_{6-x/2+\delta}$ , depends on the nature of the A ions, calcium or strontium, on the rate of substitution x, and on the oxygen pressure as shown from table III. It can

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Crystallographic data and analytical results for the oxides  $La_{2-x}A_{1+x}Cu_{2}O_{6-x/2+\delta}.$ 

L4,,,(Ca,,,,(Cu,O <sub>3,97</sub>	La:.48710.11O12O6.97 0.04	La <sub>1.9</sub> Sr <sub>1.1</sub> Cu <sub>2</sub> O <sub>1.27</sub>	LasSrCu <sub>2</sub> O <sub>4</sub>	Composition	Quenched oxides (in air)
0.02	0.04	0.02	0	<u>۰</u>	ides (i
0.02 $a=3.825 \text{ Å}$ c=19.404  Å	a= 3.959 Å c=19.956 Å	a= 3.863 Å c=19.963 Å	a = 3.865  A c = 19.887  A	Oall parameters	n ait)
La <sub>1.4</sub> Ca <sub>1.1</sub> Cu <sub>2</sub> O <sub>4.01</sub>	La1.14811.14Ct/204.94		La <sub>z</sub> SrCu <sub>1</sub> O <sub>x s</sub>	Composition	Annealed oxides (in Od)
0.08	0.29		0.20	0,	zides (i
0.08 a = 3.825 Å c=19.404 Å	0.29  a = 3.868  Å $c = 20.051  Å$	σ	a= 3.865 Å c=20.065 Å	Cell parameters	in O <sub>d</sub> )

indeed be seen for the strontium oxides synthesized in air, like La<sub>2</sub>SrCu<sub>2</sub>O<sub>6</sub>, that  $\delta$  increases with the strontium content tending towards the formulation La<sub>2</sub>\_xA<sub>1+x</sub>Cu<sub>2</sub>O<sub>6</sub>. It results that the Cu<sup>3+</sup> content increases with the divalent A ion content, in order to compensate the oxygen vacancies due to the substitution of Sr<sup>2+</sup> or Ca<sup>2+</sup> for La<sup>3+</sup>. The annealing of the latter oxides at 400° C under an oxygen pressure of one bar shows the ability of these phases to intercalate oxygen,  $\delta$  ranging from 0 to 0.29 for La<sub>2-x</sub>Sr<sub>1+x</sub>Cu<sub>2</sub>O<sub>6-x/2+\delta</sub> whereas  $0.02 \le \delta \le 0.08$  for La<sub>1.9</sub>Ca<sub>1.1</sub>Cu<sub>2</sub>O<sub>5.9\delta+\delta</sub>. One can see that the rate of intercalation is higher for the strontium oxides than for the calcium compound. Moreover it seems that in the strontium oxides the maximum rate of intercalation increases with the strontium content. Contrary to the oxides La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>1.4+\delta</sub>, the compounds La<sub>3-x</sub>Sr<sub>1+x</sub>Cu<sub>2</sub>O<sub>6-x/2+\delta</sub> exhibit a variation of the interlayer distances: the c parameter of the tetragonal cell increases with the oxygen content  $\delta$ , for a same x value. This influence of

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integrcalation on the c parameter, can be explained by the fact that the  $|\text{La}_{1}\_s\text{L}_s\text{Cu}_2\text{O}_c|_{\infty}$  slabs, which are stoichiometric and formed of SrOtype layers are more rigid than the  $|\text{Ba}_{1}\_s\text{La}_{0}\_s\text{Cu}_3\text{O}_7|_{\infty}$  slabs, and are only displaced by the introduction of oxygen between them. However the behaviour of the oxides  $\text{La}_{1}\_s\text{Ca}_{1}\_t\text{Cu}_2\text{O}_{6-s/2+\delta}$  where c parameter is independent of  $\delta$  is not explained; nevertheless in this latter case  $\delta$  remains rather weak  $(\delta \le 0.08)$ . The oxygen desintercalation of these oxides is similar to that observed for the first family: for instance heating the most oxidized compound  $\text{La}_2\text{SrCu}_2\text{O}_{6-2\delta}$  at 400° C under low oxygen pressures ( $\sim 10^{-3}$  bar) leads progressively to the reduced phase  $\text{La}_2\text{SrCu}_2\text{O}_6$ .

OXYGEN INTERCALATION IN COPPER OXIDES

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owing to the fact that the different compositions were not synthesized x contrary to La<sub>2-x</sub>Sr<sub>1+x</sub>Cu<sub>2</sub>O<sub>6-x/2+d</sub>, but increase up to  $x \simeq 1/3$  and in the case of strontium. For instance, the  $\delta$  values observed for the stronowing to the wide homogeneity ranges observed for these oxides especially tium oxides synthesized in air (table I) do not increase progressively with equilibrium is rarely reached for this series. So, for  $0 \le x \le 1$  the  $\delta$  values at the same temperature in order to obtain pure oxides. It is sure that then decrease again up to  $x\simeq 1$ . These  $\delta$  values are difficult to compare oxygen pressure. Like for the two other series, oxygen can be intercalated greater  $\delta$  values. Thus it appears that kinetics plays an important part us to prepare pure phases with the same structure but characterized by samples in the same conditions, but for longer times (24 h to 48 h) allowed given in table I correspond to heating times of 12 h and annealing these tively. The curves  $\delta = f(x)$  are given in figure 4 for the strontium comor desinterculated by annealing the samples synthesized in air, at 400°C seen that oxygen can easily be desintercalated, tending towards the most pounds where they are compared with the fine  $\delta=x/2$  which represents under an oxygen pressure of one bar or under vacuum ( $10^{-3}$  bar) respecfor oxygen intercalation in this phase at a given temperature and a given occupied in this latter composition range. From these results it seems that  $(0.33 \le x \le 1.20)$ , 11 % to 33 % of the available anionic sites being only oxygen defect structure; it appears that intercalation tends to be maximum the maximum rate of intercalation avalaible in this structure. It can be an ordering of the oxygen vacancies. Thus, rather close to the stoichiometric and LaSrCu1110, and the trend to form a related defect structure but with the trend to preserve a stoichiometric K2NiF4 structure as for La2Cu"O4 for low x values (0  $\le$  x  $\le$  0.25), whereas it is only partial for higher x values compound La2CuO4 the trend to stoichiometry is favoured by partial oxiintercalation is governed by two opposite effects which are competitive: The behaviour of the oxides La<sub>2-x</sub>A<sub>x</sub>CuO<sub>4-x/2+d</sub> is much more complex

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agreement with the observation previously made by Goodenough et al. [3]. with the rate of interculation, i. e. with the Cu3+/Cu2+ ratio, except for high x values which exhibit order-disorder phenomena. This behavior is in size of  $Sr^{2+}$  which is slightly larger than La<sup>3+</sup>. For every x value, c increases the influence of several factors: copper (III) and oxygen vacancies contents, for quenched and annealed compounds is complex (fig. 5). It results from smaller ( $c = 12.900 \, \text{A}$ ). The evolution of the c parameter versus composition « a» greater than that of the quenched specimen (a = 3.791 Å), c being an important decrease of the rate of the oxygen vacancies ( $\delta = 0.33$ ). It results that the order desappears, leading to a true tetragonal cell with 3.76 Å (table I). The annealing at 400° C in oxygen of this phase involves superstructure in the (001) plane with a «a» subcell parameter of corresponding to x=1.20. The sample quenched in air  $(\delta=0)$  exhibits a of the K2NiF4 subcell. It is for instance the case of the strontium oxide oxides, an order-disorder phenomenon of the oxygen vacancies appears in the (00 l) plane which contributes to the variation of the « a » parameter for high x values which exhibit superstructures. For such oxygen defect generally not influenced by the intercalation-desintercalation process except meter which characterizes the corresponding K2NiF4 type tetragonal cell is different microphases as observed by electron diffraction. The «a» para. more under normal oxygen pressure, and oxygen vacancies are favoured; the resulting great amount of anionic vacancies are ordered, leading to for x = 1, the stoichiometric oxide LaSrCuO<sub>4</sub> [3] cannot be stabilized any dation of Cu(II) to Cu(III), whereas rather far from La2CuO4, for example

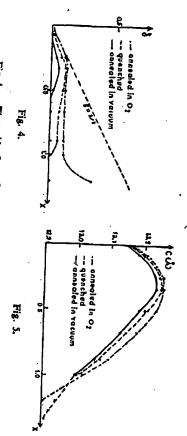


Fig. 4. — The exides  $L_{s-x}Sr_xCuO_{s-x/s+\delta}$ : evolution of  $\delta$  as a function of x for oxides resulting from different thermal treatments.

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Fig. 5. — The oxides  $L_{2r-x}SrC_{x}UQ_{4-x/3+2}$ : evolution of the c parameters a function of x for oxides resulting from different thermal treatments.

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a decrease of «c». large and its effects prevails on that of substitution Sr2+/La3+, involving and tends towards zero so that c increases owing to the replacement of For small x values (x < 0.25) the number of oxygen vacancies remains low due to substitution of Sr2+ for La3+ and decreasing due to oxygen vacancies. La<sup>3+</sup> by Sr<sup>2+</sup>. For x > 0.25 the number of oxygen vacancies becomes very The evolution of a concan be interpreted by two opposite effects: increasing

## INFLUENCE OF THE INTERCALATION PROCESS OF THE MIXED VALENCE COPPER OXIDES ON THE BLECTRICAL PROPERTIES

conductors or p type semi-metals or metals are strongly influenced by the rate of intercalation. The electron (tansport properties of these phases, which are p type semisimultaneously of Cu(II) and Cu(III), and are thus mixed-valence oxides. Most of the oxides described above are characterized by the presence

The evolution of conductivity versus reciprocal temperature for different

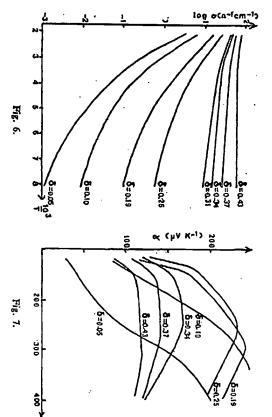


Fig. 6. — The oxides Ba, La, Cu, O14+8: variation of the conductivity (logarithmic scale) as a function of reciprocal temperature for different & values.

Fig. 7. — The oxides Ba, La, Cu, O14+3: variation of the thermoelectric power as a function of T for different & values.

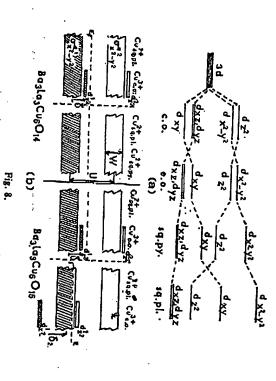
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δ values of the oxides La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>14+δ</sub> (fig. 6) shows that the conduc-

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responds to a  $dz_2^2$  empty level located just above or accross the filled to a semi-metallic or metallic conduction for the limit Ba3La3Cu6O15 corsynthesized; on the other hand, the only level configuration which can lead of La<sub>3</sub>Ba<sub>3</sub>Cu<sub>6</sub>O<sub>1.4</sub> (fig. 8 b) is that of an insulator but this limit has not been few eV like the  $\sigma_{x_1-x_2}^{\bullet t}$  and  $\sigma_{x_2-x_2}^{*2}$  bands. It results that the band structure strong electron-electron interactions split the  $dz_2^1$  and  $dz_2^2$  levels by a bons along c. The  $\sigma_{r_2-r_2}^2$  bands result from Cu - O - Cu interactions and and the oxidized form Ba<sub>3</sub>La<sub>3</sub>Cu<sub>6</sub>O<sub>13</sub> which exhibits infinite octahedral ribby ribbons of one octahedron and two tetragonal pyramids running along c, mixing of the two limits: the reduced form Ba<sub>2</sub>La<sub>2</sub>Cu<sub>6</sub>O<sub>14</sub> characterized crystal field [15] (fig. 8 a). Every composition can indeed be considered as a guration is mainly determined by the splitting of the 3d Cu orbitals by the power of these phases (fig. 7) is very sensitive to the intercalation rate. These properties are interpreted by a conduction band model whose confithe structure which remains unchanged. In the same way the thermoelectric tivity increases drastically with the intercalation of oxygen, contrary to



b) Schematic band diagram for Ba, La, Cu,O1, and Ba, La, Ou,O1. U donotes the intrad) Crystal field splitting for d element in different environments; c. o.: compressed octaoctahedrs and W the estimated band width. atomic coulomb energy,  $\delta$ ,  $\delta_1$  and  $\delta_2$  the splitting due to the axial distortion of the hedron, e. o.: elongated octahedron, sq. py.: square pyramid, sq. pl. square plane.

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state can be explained by the Mott model [16] of quasi localized holes corresponds to a local change of copper coordination, will involve an trapped at the top of the filled  $3 d_{x^{1-y^2}}$  band.  $\sigma_{x^{1}-y^{1}}^{*}$  band (fig. 8 b). Thus, it appears that intercalation of oxygen which This progressive transition from a semi-conductive to a semi-metallic linear evolution of log  $\sigma$  vs  $\delta$  at 293 K is in agreement with this model increase of the number of holes in the conduction band. The approximately increase of the density of the  $dz_2^2$  levels above the filled  $\sigma_{x^2-r^2}^{*1}$  band, i. e. an

confirm this influence of intercalation:  $\alpha$  increases continuously with T for  $La_{1.90}Sr_{1.10}Cu_2O_{5.97}$ . The Seebeck coefficient curves  $\alpha = f(T)$  (fig. drastically the c parameter: the calcium oxide La<sub>1.90</sub>Ca<sub>1.10</sub>Cu<sub>2</sub>O<sub>3.97</sub> is conductive to a semi-metallic state is observed as the oxygen intercalation rate indeed much more conductor than the corresponding strontium oxide The conductivity depends also on the nature of the A ion which influences increases from  $\delta = 0$  (La<sub>2</sub>SrCu<sub>2</sub>O<sub>6</sub>) to  $\delta = 0.29$  (La<sub>1.86</sub>Sr<sub>1.14</sub>Cu<sub>2</sub>O<sub>6.22</sub>). 300 K (fig. 9) it can be seen that a continuous transition from a semi-From the evolution of the curves  $\log \sigma = f(1/T)$ , between 80 K and The oxides La<sub>2-x</sub>A<sub>1+x</sub>Cu<sub>2</sub>O<sub>6-x/2+8</sub> exhibit a similar behaviour [17]

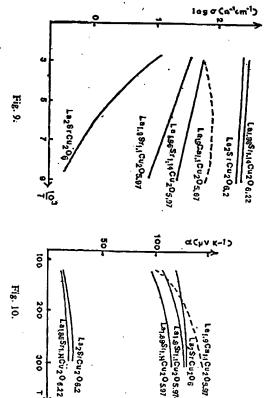


Fig. 9. — The exides  $\text{La}_{\ell-x}A_{1+x}\text{Cu}_1\text{O}_{\ell-x/\ell+3}$ ; evolution of the conductivity (logarithmic scale) vs  $T^{-1}$  for different compositions.

evolution of the thermoelectric power vs T for different compositions. Fig. 10. — The oxides Laz-A1+xC10-x/+8:

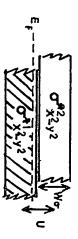
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OXYGEN INTERCALATION IN COPPER OXIDES

C. NICHEL AND B. RAVEAU

gives rise to a higher mobility. the calcium compound so that the overlapping of the two  $\sigma_{x^{2}-y^{2}}$  bands strontium oxide shows that the band width  $W_{\sigma}$  must be larger than U in of the calcium oxide La<sub>1.90</sub>Ca<sub>0.10</sub>Cu<sub>2</sub>O<sub>3.97</sub> compared to the corresponding W, (fig. 11). In the same manner the relatively high and metallic conductivity energy U is in this case of the same order of magnitude as the band width of the very weak  $Cu^{3+}$  content— $\delta \simeq 0$ —let us think that the intra-atomic مَيْمُ band. However the rather high conductivity of La<sub>2</sub>SrCu<sub>2</sub>O<sub>6</sub> in spite be explained by the same Mott model of holes trapped at the top of the high intercalation rates ( $\sigma = 0.20$  to 0.29), i. e. for high hole concentration These properties very similar to those obtained for La3Ba3Cu6O14+6 can whereas it becomes weak and nearly independent of the temperature, for the small intercalation rates ( $\delta = 0$  to 0.04) *l. e.* for small Cu<sup>3+</sup> contents



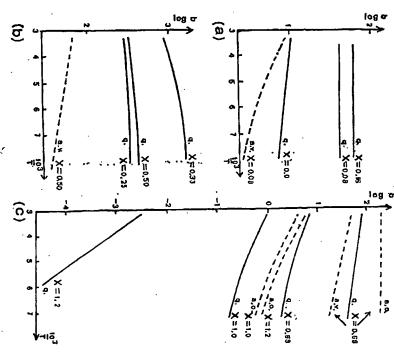
schematic band diagram as deduced from electron transport properties Fig. 1L - The oxides Lag-xAg+xCugOq-x/g+8:

also confirmed by the fact that a increases with temperature (fig. 13). The agreement with this model. The fact that the holes may be trapped on locathe thermoelectric power values (fig. 13) greater than those of a metal are in not correspond to the metallic model  $\rho = \rho_0(1 + \gamma t)$  (fig. 12 a), as well as developped by Goodenough for La2CuO4 [19] involving the presence of semi-metallic behaviour and their properties can be interpreted by the model and  $0.50 < x \le 1.20$  for the oxides quenched in air and annealed in oxygen. domains must in fact be distinguished:  $0 \le x \le 0.16$ ,  $0.16 < x \le 0.50$ fized levels at the top of the  $\sigma_{x+x}^{s}$  band according to the Mott Model is holes in the filled band  $\sigma_{x_1,x_2}^{x_1}$ . The weak variation of conductivity which does The compounds of the first domain  $(0 \le x \le 0.16)$  are characterized by a factor governing the electron transport properties of the phases. Three as  $\alpha = f(T)$  is more complex than the two other series:  $\delta$  is not the only ranging from 80 K to 300 K. However the evolution of log  $\sigma$  vs 1/T as well with the rate of intercalation o as shown from figure 12 for temperature  $CuO_{4-\pi/2+\delta}$  [18]. For a given substitution rate x, the conductivity increases The highest conductivities are observed for the oxides La<sub>2-x</sub>Sr,

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a)  $0 \le x \le 0.16$ ; b)  $0.16 < x \le 0.50$ ; c)  $0.5 < x \le 1.20$ Fig. 12.—The oxides La<sub>2-3</sub>Sr<sub>2</sub>CuO<sub>4-3/14-8</sub>: variation of the conductivity vs T<sup>-1</sup> (g: quenched in air, a. o.: annealed in O<sub>2</sub>, a. v.: annealed in vacuum).

conductivity (fig. 12 b) which increases with the intercalation rate: ho increases (0.50 <  $x \le 1.20$ ), exhibit for the less oxidized compounds synthetized in oxides belonging to the second domain (0.16  $< x \le 0.50$ ), exhibit a metallic relation  $\sigma = A \exp \left[-(Q/k_0T)^{1/4}\right]$  which characterizes a variable range hopp δ (fig. 12 c), and correlatively α increnses as δ decreases (fig. 13). These air ( $\delta < 0.07$ ) a semi-conductive behaviour:  $\sigma$  decreases drastically with and nearly temperature independent (fig. 13). The highest x compositions I and 1.2 exhibit a variation of the conductivity according to the Mott latter oxides and especially the compositions corresponding to x=0.88, linearly with temperature and the thermoelectric power values are weak

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I bar exhibit the same  $\delta$  value ( $\delta \simeq 0.11$ ), but are characterized by a decrease to  $0.33 \le x \le 1.0$  which have been annealed under an oxygen pressure of do not depend on the  $\delta$  value only. So, for instance, the oxides corresponding ing of holes located in the  $\sigma_{x^2-y^2}^{*1}$  band close to the Fermi level. It must be noted that the electrical properties of the oxides La2-\_Sr\_CuO4-x/2+&

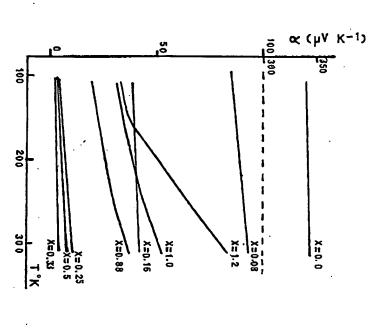


Fig. 13. — The oxides La<sub>1-x</sub>Sr<sub>x</sub>CuO<sub>1-xl1+8</sub>: thermoelectric power vs T for quenched oxides with different x values.

of anionic vacancies  $(x/2-\delta)$  on the carrier mobility. Moreover the distriinfluence the electron transport properties of these compounds. bution of the oxygen defects i. e.; the order-disorder phenomena, may of  $\sigma$  as x increases as shown figure 14. This shows the influence of the rate

owing to the interculation or desinterculation of oxygen. For this reason electrical conductivity can vary drastically, under a given oxygen pressure The great sensitivity of these compounds to oxygen makes that their

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results from desintercalation and intercalation of oxygen successively. Similar sively by a weight loss and weight gain, show clearly that this behaviour 650 K. The thermogravimetric curves of these phases, characterized success range 300 K-420 K and then increases again in the temperature range 420 Koxygen pressure of 0.2 bar that  $\sigma$  decreases first drastically in the temperature air  $(0 \le \delta \le 0.04)$ . One indeed observes (fig. 15), beyond 300 K under an Such anomalies of the conductivity have indeed been observed for the oxides of the three families are not sensitive to intercalation or desintercalation at relatively low temperatures (T < 300 K), where all the compounds we have only discussed above the electrical properties of these phases evolution of the conductivity of La<sub>1.9</sub>Ca<sub>1.1</sub>Cu<sub>2</sub>O<sub>5.97</sub> versus reciproca reversibility of the intercalation process in these phases is illustrated by the measurements confirm the oxygen desintercalation-intercalation process. The properties are observed for the oxides  $\text{La}_{2-x}\text{Sr}_{1+x}\text{Cu}_2\text{O}_{6-x/2+\delta}$  synthesized  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4-x/2+\delta}$  corresponding to  $0 \le x \le 0.16$  and synthesized in in air for x = 0.1 and 0.14 (fig. 15) and for which the thermogravimetric

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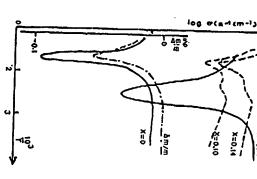
OXYGEN INTERCALATION IN COPPER OXIDES

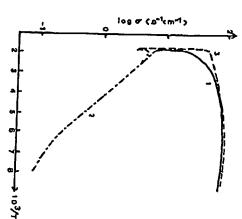
x=0.50

\$=0.89

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example to illustrate the close relation between variation of conductivity and oxygen La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>1-x/7+8</sub> recies (solid line) and in the La<sub>2-x</sub>Sr<sub>1+x</sub>Cu<sub>2</sub>O<sub>3-x1/+8</sub> series (dolted line). A TG curve for x=0 (first series), with the same temperature scale is given as - Variation of conductivity vs T-4(T > 300K) in air for some oxides in the

Fig. 16.

Fig. 16. — Variation of conductivity vs T-1 for the oxides La1.9Ca1.1Cu1O1.41 under different atmospheres:

- first heating under inert atmosphere,
- じと
- --- first cooling and second heating under inert atmosphere air introduction and second cooling (in air).

Heating again up to 500 K under argon leads to the same curve. However tive behaviour is observed owing to the lower oxygen rate of intercalation. progressively down to 77 K. In this latter temperature range a semi-conducthis stage of the experiment, heating is stopped and the sample is cooled the departure of oxygen and at about 570 K  $\sigma$  decreases drastically. At under argon as soon as the temperature is greater than 300 K owing to temperature under argon and air (fig. 16). The behaviour of this phase indeed very different in argon and in air. The conductivity decreases

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# OXYGEN INTERCALATION IN COPPER OXIDES

at 570 K. At 570 K argon is replaced by air, and heating is stopped. It can the thermodynamical equilibrium is not yet reached when we stop heating that observed for the starting material intercalation. The behaviour observed from 570 K to 77 K is then similar to beyond 500 K, or decreases again; this phenomenon is due to the fact that that the conductivity increases immediately owing to the oxygen

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(Received December 12, 1983)

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#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of

J. Bednorz et al.

Date: December 15, 1998

Serial No. 08/303,561

Group Art Unit: 1105

Filed: September 9, 1994

Examiner: M. Kopec

For:

NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE, AND METHODS FOR THEIR USE AND PREPARATION

#### AFFIDAVIT UNDER 37 C.F.R. 1.132

Commissioner of Patents and Trademarks Washington, D. C. 20231

Sir:

I, David B. Mitzi, being duly sworn, do hereby depose and state:

That I received a B. S. E. degree in Electrical Engineering/Engineering Physics (1985) from Princeton University and a PhD. degree, in Applied Physics (1990) from Stanford University, California.

That I have worked as a research staff member in Solid State Chemistry at the Thomas Watson Research Center of the International Business Machines Corporation in Yorktown Heights, NY from 1990 to the present.

That I have worked in the fabrication of and characterization of high temperature superconductor and related materials from 1990 to the present.

That I have reviewed the above-identified patent application and that I have reviewed the above-identified patent application and acknowledge that it represents the work of Bednorz and

Muller, which is generally recognized as the first discovery of superconductivity above 26°K and that subsequent developments in this field have been based on this work.

That all the high temperature superconductors which have been developed based on the work of Bednorz and Muller behave in a similar manner, conduct current in a similar manner and have similar magnetic properties.

That once a person of skill in the art knows of a specific transition metal oxide composition which is superconducting above 26°K, such a person of skill in the art, using the techniques described in the above-identified patent application, which includes all knows principles of ceramic fabrication known at the time the application was filed, can make the transition metal oxide compositions encomposed by the claims in the above identified application, without undue experimentation or without requiring ingenuity beyond that expected of a person of skill in the art. This is why the work of Bednorz and Muller was reproduced so quickly after their discovery and why so much additional work was done in this field within a short period of their discovery.

The general principles of ceramic science referred to by Bednorz and Mueller in their patent application can be found in many books and articles published before their discovery. An exemplary list of books describing the general principles of ceramic fabrication are:

- 1) Introduction to Ceramics, Kingery et al., Second Edition, John Wiley & Sons, 1976, in particular pages 5-20, 269-319, 381-447 and 448-513, a copy of which is with the Affidavit of Thomas Shaw submitted December 15, 1998.
- 2) Polar Dielectrics and Their Applications, Burfoot et al., University of California Press, 1979, in particular pages 13-33, a copy of which is with the Affidavit of Thomas Shaw submitted December 15, 1998.
- 3) Ceramic Processing Before Firing, Onoda et al., John Wiley & Sons, 1978, the entire book, a copy of which is with the Affidavit of Thomas Shaw submitted December 15, 1998.

4) Structure, Properties and Preparation of Perovskite-Type Compounds, F.S. Glasso, Pergamon Press, 1969, in particular pages 159-186, a copy of which is with the Affidavit of Thomas Shaw submitted December 15, 1998.

An exemplary list of articles applying their general principles of ceramic fabrication to the types of materials described in applicants' specification are (these references are cited on applicant's 1449 form submitted August 5, 1987 and in PTO Form 892 in Paper # 20, Examiner's action dated August 8, 1990):

- 1) Oxygen Defect K<sub>2</sub>NiF<sub>4</sub> Type Oxides: The Compounds La<sub>2-x</sub> Sr<sub>x</sub>CuO<sub>4-x/2+\*</sub>, Nguyen et al., Journal of Solid State Chemistry 39, 120-127 (1981).
- 2) The Oxygen Defect Perovskite BaLa<sub>4</sub> Cu<sub>5</sub>  $\bigcirc_{13.4}^{0}$ , A Metallic Conductor, C. Michel et al., Mat. Res. Bull., Vol. 20, pp. 667-671, 1985.
- 3) Oxygen intercalation in mixed valence copper oxides related to the perovskite, C. Michel et al., Revue de Chemie minerale, p. 407, 1984.
- 4) Thermal Behaviour of Compositions in the Systems x BaTiO<sub>3</sub> + (1-x) Ba(Ln<sub>0.5</sub> B<sub>0.5</sub>) 0<sub>3</sub>, V.S. Chincholkar et al. Therm. Anal. 6th, Vol. 2., p. 251-6, 1980.

David B. Mitzi

Sworn to before me this 15 th day of Decemp

**Notary Public** 

DANIEL P. MORRIS NOTARY PUBLIC, State of New York No. 4888676 Qualified in Westchester County

Commission Expires March 16, 19

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Examiner: M. Kopec

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TEMPERATURE, AND METHODS FOR THEIR USE AND PREPARATION

#### AFFIDAVIT UNDER 37 C.F.R. 1.132

Commissioner of Patents and Trademarks Washington, D. C. 20231

Sir:

I, Timothy Dinger, being duly sworn, do hereby depose and state:

That I received a B. S. degree in Ceramic Engineering (1981) from New York State College of Ceramics, Alfred University, an M. S. degree (1983) and a PhD. degree (1986), both in Material Science from the University of California at Berkley.

That I have worked as a research staff member in Material Science at the Thomas Watson Research Center of the International Business Machines Corporation in Yorktown Heights, NY from 1986 to the present.

That I have worked in the fabrication of and characterization of high temperature superconductor materials from 1987 to 1991

That I have reviewed the above-identified patent application and acknowledge that it represents the work of Bednorz and Muller, which is generally recognized as the first discovery of

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That all the high temperature superconductors which have been developed based on the work of Bednorz and Muller behave in a similar way, conduct current in a similar manner and have similar magnetic properties.

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Sworn to before me this 16th day of Sommer

Notary Public

SANDRA M. EMMA Notary Public, State of New York No. 01PO4935290 Qualified in Westchester County Commission Expires July 5, 🕰



#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of

J. Bednorz et al.

Date: December 15, 1998

Serial No. 08/303,561

Group Art Unit: 1105

Filed: September 9, 1994

Examiner: M. Kopec

For:

NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE, AND METHODS FOR THEIR USE AND PREPARATION

#### AFFIDAVIT UNDER 37 C.F.R. 1.132

Commissioner of Patents and Trademarks Washington, D. C. 20231

Sir:

I, Chang C. Tsuei, being duly sworn, do hereby depose and state:

That I received a B. S. degree in Mechanical Engineering from National Taiwan University (1960) and M. S. and PhD. degrees, in Material Science (1963, 1966) respectively from California Institute of Technology.

That I have worked as a research staff member and manager in the physics of superconducting, amorphous and structured materials at the Thomas Watson Research Center of the International Business Machines Corporation in Yorktown Heights, New York from 1973 to the present. (See attached Exhibit A for other professional employment history.)

That I have worked in the fabrication of and characterization of high temperature superconductor and related materials from 1973 to the present.

That I have reviewed the above-identified patent application and acknowledge that it represents the work of Bednorz and Muller, which is generally recognized as the first discovery of YO987-074BY

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- 3) Ceramic Processing Before Firing, Onoda et al., John Wiley & Sons, 1978, the entire book, a copy of which is with the Affidavit of Thomas Shaw submitted December 15, 1998.

4) Structure, Properties and Preparation of Perovskite-Type Compounds, F.S. Glasso, Pergamon Press, 1969, in particular pages 159-186, a copy of which is with the Affidavit of Thomas Shaw submitted December 15, 1998.

An exemplary list of articles applying their general principles of ceramic fabrication to the types of materials described in applicants' specification are (these references are cited on applicant's 1449 form submitted August 5, 1987 and in PTO Form 892 in Paper # 20, Examiner's action dated August 8, 1990):

- 1) Oxygen Defect K<sub>2</sub>NiF<sub>4</sub> Type Oxides: The Compounds La<sub>2-x</sub> Sr<sub>x</sub>CuO<sub>4-x/2+\*</sub>, Nguyen et al., Journal of Solid State Chemistry 39, 120-127 (1981).
- 2) The Oxygen Defect Perovskite BaLa<sub>4</sub> Cu<sub>5</sub>-0<sub>13.4</sub>, A Metallic Conductor, C. Michel et al., Mat. Res. Bull., Vol. 20, pp. 667-671, 1985.
- 3) Oxygen intercalation in mixed valence copper oxides related to the perovskite, C. Michel et al., Revue de Chemie minerale, p. 407, 1984.
- 4) Thermal Behaviour of Compositions in the Systems x BaTiO<sub>3</sub> + (1-x) Ba(Ln<sub>0.5</sub> B<sub>0.5</sub>) 03, V.S. Chincholkar et al. Therm. Anal. 6th, Vol. 2., p. 251-6, 1980.

Sworn to before me this 16 th day of Vecentur

**Notary Public** 

SANDRA M. EMMA Notary Public, State of New York No. 01PO4935290 Qualified in Westchester Count Commission Expires July 5,

#### CHANG C. TSUEI

#### **Education**

California Institute of Technology, M.S. (1963), Ph.D. (1966) National Taiwan University, B.S. (1960)

#### Professional Employment

1993 - present - Research Staff Member

1983 - 1993 - Manager, Physics of Structured Materials

1979 - 1983 - Manager, Physics of Amorphous Materials

1974 - 1975 - Acting Manager, Superconductivity

1973 - 1979 - Research Staff Member

Harvard University: 1980 (Summer)

Visiting Scholar in Applied Physics

Stanford University: 1982 (Sept.) - 1983 (April)

Visiting Scholar in Applied Physics

#### California Institute of Technology

1972 - 1973 - Senior Research Associate in Applied Physics

1969 - 1972 - Senior Research Fellow in Materials Science

1966 - 1969 - Research Fellow in Materials Science

Exhibit A

#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J. Bednorz et al.

Date: December 15, 1998

Serial No. 08/303,561

Group Art Unit: 1105

Filed: September 9, 1994

Examiner: M. Kopec

For: NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE, AND METHODS FOR THEIR USE AND PREPARATION

The Commissioner of Patents and Trademarks Washington, D.C. 20231

#### **AFFIDAVIT UNDER 37 CFR 1.132**

Sir:

I, Thomas M. Shaw, being duly sworn, do hereby depose and state:

I received a B.S. degree in Metallurgy from the University of LIverpool, Liverpool, England and a M.S. and PhD. degree in Materials Science (1981) from the University of California, Berkeley.

I have worked as a postdoctoral researcher in the Material Science Department of Cornell University from 1981-1982. I worked at Rockwell International Science Center in Thousand Oaks, California from 1982-1984 as a ceramic scientist. I have worked as a research staff member in Ceramics Science at the Thomas J. Watson Research

Center of the International Business Machines Corporation in Yorktown Heights, N.Y. from 1984 to the present.

I have worked in the fabrication of and characterization of ceramic materials of various types, including superconductors and related materials from 1984 to the present.

Attached is a resume of my publications. I have reviewed the above-identified patent application and acknowledge that it represents the work of Bednorz and Mueller, which is generally recognized as the first discovery of superconductivity above 26°K and that subsequent developments in this field have been based on this work.

That all the high temperature superconductors which have been developed based on the work of Bednorz and Mueller behave in a similar manner, conduct current in a similar manner and have similar magnetic properties.

That once a person of skill in the art knows of a specific transition metal oxide composition which is superconducting above 26°K, such a person of skill in the art, using the techniques described in the above-identified patent application, which includes all known principles of ceramic fabrication known at the time the application was filed, can make the transition metal oxide compositions encompassed by the claims in the above-identified application, without undue experimentation or without requiring ingenuity beyond that expected of a person of skill in the art. This is why the

work of Bednorz and Mueller was reproduced so quickly after their discovery and why so much additional work was done in this field within a short period of their discovery.

The general principles of ceramic science referred to by Bednorz and Mueller in their patent application can be found in many books and articles published before their discovery. An exemplary list of books describing the general principles of ceramic fabrication are:

- 1) Introduction to Ceramics, Kingery et al., Second Edition, John Wiley & Sons, 1976, in particular pages 5-20, 269-319, 381-447 and 448-513, a copy of which is attached herewith.
- 2) Polar Dielectrics and Their Applications, Burfoot et al., University of California Press, 1979, in particular pages 13-33, a copy of which is attached herewith.
- 3) Ceramic Processing Before Firing, Onoda et al., John Wiley & Sons, 1978, the entire book, a copy of which is attached herewith.
- 4) Structure, Properties and Preparation of Perovskite-Type Compounds, F.S. Glasso, Pergamon Press, 1969, in particular pages 159-186, a copy of which is attached herewith.

An exemplary list of articles applying their general principles of ceramic fabrication to the types of materials described in applicants' specification are (these references are cited on applicant's 1449 form submitted August 5, 1987 and in PTO Form 892 in Paper # 20, Examiner's action dated August 8, 1990):

- 1) Oxygen Defect K<sub>2</sub>NiF<sub>4</sub> Type Oxides: The Compounds La<sub>2-x</sub> Sr<sub>x</sub> CuO<sub>4-x/2+δ</sub>, Nguyen et al., Journal of Solid State Chemistry 39, 120-127 (1981).
- 2) The Oxygen Defect Perovskite BaLa₄ Cu₅-0₁₃₄, A Metallic Conductor , C. Michel et al., Mat. Res. Bull., Vol. 20, pp. 667-671, 1985.

- 3) Oxygen intercalation in mixed valence copper oxides related to the perovskite, C. Michel et al., Revue de Chemie minerale, p. 407, 1984.
- 4) Thermal Behaviour of Compositions in the Systems x BaTiO<sub>3</sub> + (1-x) Ba(Ln<sub>0.5</sub> B<sub>0.5</sub>)  $0_3$ , V.S. Chincholkar et al. Therm. Anal. 6th, Vol. 2., p. 251-6, 1980.

Bv:	Sfromas M. Sligar	^
	Thomas M. Shaw	

Sworn to before me this	11.14h		Do no so los os	1998
Sworn to before me this	172	_day of _	William C	 

Notary Public

SANDRA M. EMMA
Notary Public, State of New York
No. 01PO4935290
Qualified in Westchester County
Commission Expires July 5.

#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J. Bednorz et al.

Date: December 18, 1998

Serial No. 08/303,561

Group Art Unit: 1105

Filed: September 9, 1994

Examiner: M. Kopec

For: NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE, AND METHODS FOR THEIR

**USE AND PREPARATION** 

The Commissioner of Patents and Trademarks Washington, D.C. 20231

#### **AFFIDAVIT UNDER 37 CFR 1.132**

Sir:

I, Peter R. Duncombe, being duly sworn, do hereby depose and state:

I received a B.A. degree in Chemistry from the State University of New York at New Paltz, New Paltz, N.Y. and a M.S. degree in Chemical Engineering (1983) from the State University of New York at Buffalo, Buffalo, N.Y.

I have worked as a graduate research assistant in the Chemical Engineering

Department of SUNY at Buffalo from 1980-1983. I have worked as a chemical
engineer in Ceramics Science at the Thomas J. Watson Research Center of the
International Business Machines Corporation in Yorktown Heights, N.Y. from 1984 to
the present.

I have worked in the fabrication of and characterization of ceramic materials of various types, including superconductors and related materials from 1984 to the present.

Attached is a resume of my publications (Attachment A).

I have reviewed the above-identified patent application and acknowledge that it represents the work of Bednorz and Mueller, which is generally recognized as the first discovery of superconductivity above 26°K and that subsequent developments in this field have been based on this work.

That all the high temperature superconductors which have been developed based on the work of Bednorz and Mueller behave in a similar manner, conduct current in a similar manner and have similar magnetic properties.

That once a person of skill in the art knows of a specific transition metal oxide composition which is superconducting above 26°K, such a person of skill in the art, using the techniques described in the above-identified patent application, which includes all known principles of ceramic fabrication known at the time the application was filed, can make the transition metal oxide compositions encompassed by the claims in the above-identified application, without undue experimentation or without requiring ingenuity beyond that expected of a person of skill in the art. This is why the

work of Bednorz and Mueller was reproduced so quickly after their discovery and why so much additional work was done in this field within a short period of their discovery.

The general principles of ceramic science referred to by Bednorz and Mueller in their patent application can be found in many books and articles published before their discovery. An exemplary list of books describing the general principles of ceramic fabrication are:

- 1) Introduction to Ceramics, Kingery et al., Second Edition, John Wiley & Sons, 1976, in particular pages 5-20, 269-319, 381-447 and 448-513, a copy of which is attached herewith.
- 2) Polar Dielectrics and Their Applications, Burfoot et al., University of California Press, 1979, in particular pages 13-33, a copy of which is attached herewith.
- 3) Ceramic Processing Before Firing, Onoda et al., John Wiley & Sons, 1978, the entire book, a copy of which is attached herewith.
- 4) Structure, Properties and Preparation of Perovskite-Type Compounds, F.S. Glasso, Pergamon Press, 1969, in particular pages 159-181, a copy of which is attached herewith.

An exemplary list of articles applying their general principles of ceramic fabrication to the types of materials described in applicants' specification are (these references are cited on applicant's 1449 form submitted August 5, 1987 and in PTO Form 892 in Paper # 20, Examiner's action dated August 8, 1990):

- 1) Oxygen Defect K₂NiF₄ Type Oxides: The Compounds La₂-x Srx CuO₄-x/2+δ, Nguyen et al., Journal of Solid State Chemistry 39, 120-127 (1981).
- 2) The Oxygen Defect Perovskite BaLa₄ Cu₅-0₁₃₄, A Metallic Conductor , C. Michel et al., Mat. Res. Bull., Vol. 20, pp. 667-671, 1985.

- 3) Oxygen intercalation in mixed valence copper oxides related to the perovskite, C. Michel et al., Revue de Chemie minerale, p. 407, 1984.
- 4) Thermal Behaviour of Compositions in the Systems x BaTiO<sub>3</sub> + (1-x) Ba( $Ln_{0.5}$  B<sub>0.5</sub>) O<sub>3</sub>, V.S. Chincholkar et al. Therm. Anal. 6th, Vol. 2., p. 251-6, 1980.

I have recorded research notes relating to superconductor oxide (perovskite) compounds in technical notebook IV with entries from November 12, 1987 to June 14, 1988 and in technical notebook V with entries continuing from June 7, 1988 to May 2, 1989. Complete copies of each of these notebooks are attached - Attachment B - Book IV and Attachment C - Book V. Below is a listing of some of the compounds I prepared and recorded in these notebooks according to the teaching as described in the Bednorz and Mueller patent application using the general principles of ceramic science as described in the books and articles listed above.

In Book IV, Y<sub>1</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> batch C1 pellet pressing, sintering notes and powder processing specifications start on page 2 and continue intermittently to pg. 40 (pg. 13 has superconductive susceptibility curves for pellet 9). Batch C2 Y<sub>1</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>3</sub> detailed from pages 14 to 47.

In Book V green phase (Y<sub>2</sub>BaCuO<sub>x</sub>) microstructural photomicrographs are logged on pages 15-17 with notes continuing to pg. 19. The perovskite superconductor BiSrCaCu oxide (Bi<sub>2.15</sub>Sr<sub>1.68</sub>Ca<sub>1.7</sub>Cu<sub>2</sub>O<sub>8+8</sub>) and related perovskites Ca<sub>(2-x)</sub>Sr<sub>x</sub>CuO<sub>x</sub> and Bi<sub>2</sub>Sr<sub>2</sub>CuO<sub>x</sub> synthesis notations start and continue through pg. 61 with microstructural photomicrographs.

A series of Y<sub>1</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> stoichiometric perturbations to study compositional effects on 2nd phase or grain boundary phases and their effect on conductivity (resistivity), sintering behavior etc., continue until the end of the book notes on the page dated May 2, 1989 (page not numbered). These are typical perovskite synthetic procedures, microstructural photomicrographs, powder processing methods, characteristic susceptibility curve(s), sintering behavior and the like. Additional notes may be available in later notebooks.

The undersigned affiant swears further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or patent issuing thereon.

Sworn to before me this

day of

Olcember

19 78.

Notary Public

SANDRA M. EMMA
Notary Public, State of New York
No. 01PO4935290
Qualified in Westchester County
Commission Expires July 5, 2000

#### ATTACHMENT A

- Compensation doping of Ba0.7Sr0.3TiO3 thin films
   Copel, M Baniecki, JD Duncombe, PR Kotecki, D
   Laibowitz, R Neumayer, DA Shaw, TM
   APPLIED PHYSICS LETTERS V73 N13 SEP 28 1998 P1832-1834
- Method for Forming Noble Metal Oxides and Structures Formed Thereof. June 1998.
   Duncombe, P. R. Hummel, J. P. Laibowitz, R. B.
   Neumayer, D. A. Saenger, K. L. Schrott, A. G.
   RC 98A 41575
- Growth of Bismuth Titanate Films By Chemical Vapor Deposition and Chemical Solution Deposition. March 1998. RC-21124 Neumayer, D. A. Duncombe, P. R. Laibowitz, R. B. Shaw, T. Purtell, R. Grill, A.
- Dielectric relaxation of Ba0.7Sr0.3TiO3 thin films from 1 mHz to 20 GHz Baniecki, JD Laibowitz, RB Shaw, TM Duncombe, PR Neumayer, DA Kotecki, DE Shen, H Ma, QY APPLIED PHYSICS LETTERS V72 N4 JAN 26 1998 P498-500
- Contrasting magnetic and structural properties of two La manganites with the same doping levels McGuire, T.R. Duncombe, P.R. Gong, G.Q. Gupta, A. Li, X.W. Pickart, S.J. Crow, M.L. J. Appl. Phys. (USA) Vol.83, No.11 1 June 1998 P7076-8
- Effects of Annealing Conditions on Charge Loss Mechanisms in MOCVD (Ba0.7,Sr0.3)TiO3
   Thin Film Capacitors.
   Baniecki, J.D., Laibowitz, RB Shaw, TM Duncombe, PR Saenger, KL Cabral C
   Kotecki, DE, Shen, H, Lian, J., Ma, QY
- 7. Low Operating Voltage and High Mobility Field Effect Transistors Comproising Pentacene and Relatively High Dielectric Constant Insulators RC21233(94806) 7/17/98 Dimitrakopoulos, CD Purushothaman S, Kymissis J. Callegari A., Neumayer DA, Duncombe PR, Laibowitz RB, Shaw JM
- Maximum Magnetorsistance in Granular Manganite/Insulator System close to Percolation Threshold PACS 10/06/98
   DK Petrov, L Krusin-Elbaum, JZ Sun, C Feild, & PR Duncombe
- 9. Magnetorsistance and Hall Effect of Chromium Dioxide Epitaxial Thin Films X.W. Li, A. Gupta, T.R. McGuire, P.R. Duncombe, Gang Xiao
- Progress Report on High-k dielectric material: amorphous BST from solgel (09/98)
   P. Andry, D. Neumayer, P. Duncombe, C. Dimitrakopoulos, F. Libsch, A. Grill, R. Wisnieff

RC21352 (96175) 2 Dec1998

#### InfoGate from The IBM Total Information Retrieval Center



#### INCOMPLETE

#### **Personal Inventor History**

Loc: RES YORKTOWN Name:Duncombe, P.R. Serial:155139 Patent Pts:36 TDB Pts:1 Total Pts:37 Plateau Lvl:3 Plateau Date:10/24/98 File Update:11/02/98 Awards Due: None Title: NOVEL METAL ALKOXYALKOXIDECARBOXYLATES AND USE TO FORM FILMS Status:Filed 06/17/98 Opened as Discl Y08980231 Action: File 06/22/98 Discl Review 09/04/98 Filed as Docket Y0998254 in US Rating: 2 Pts:3 Co-inventors: Neumayer, D.A. Title: SELECTIVE GROWTH OF FERROMAGNETIC FILMS FOR MAGNETIC MEMORY, STORAGE-BASED DEVICES, AND OTHER DEVICES Status:Filed 06/17/98 Opened as Discl Y08980225 Action: File 06/29/98 Discl Review 10/15/98 Filed as Docket Y0998268 in US Pts:3 Rating: 2 Co-inventors: Guna, S. Gupta, A. Bojarczuk, N.A. Karasinski, J.M. Title: BEOL DECOUPLING CAPACITOR MATERIALS Status:Opened 01/28/98 Opened as Discl Y08980024 in US Action: File 06/24/98 Discl Review Co-inventors: Rosenberg, R. Ning, T.H. Shaw, T.M. Edelstein, D.C. Neumayer, D.A. "FABRICATION OF STOUTIUM BISMITH TINTALETE BISMITH THOMASE MUTTINGE FERROELECTRIC Laibowitz, R.B. 10/01/97 Opened as Discl Y08970512 in US Status:Opened Action: File 09/16/98 Discl Review 10/30/98 SENT TO CONSEL(L. Schuke) Title: CAPACITORS WITH AMORPHOUS DIELECTRICS AND IMPROVED DIELECTRIC PROPERTIES MADE USING SILICON SURFACES AS ELECTRODES 06/06/97 Opened as Discl Y08970261 in US Status:Opened Co-inventors: Shaw, T.M. Neumayer, D.A. Laibowitz, R.B. Title: FABRICATION OF THIN FILM FIELD EFFECT TRANSISTOR COMPRISING AN ORGANIC SEMICONDUCTOR AND CHEMICAL SOLUTION DEPOSITED METAL OXIDE Status: Filed 03/25/97 Opened as Discl Y08970113 Action: File 03/25/97 Discl Review Rating: 2 Pts:3 03/25/97 Filed as Docket Y0997083 in US 03/24/98 Filed as Docket Y0997083 in JA Rating: 2 Rating: 2 03/16/98 Filed as Docket Y0997083 in TA 03/12/98 Filed as Docket Y0997083 in KO Rating: 2 04/24/98 Last Office Action Co-inventors: Purushothaman, S. Dimitrakopoulos, C.D. Furman, B.K. Neumayer, D.A. Laibowitz, R.B.

Title: NOVEL ALKOXYALKOXIDES AND USE TO FORM FILMS

10/30/96 Opened as Discl Y08960411 Status: Filed

03/10/97 Disc1 Review Action: File

(5) 01/30/98 Filed as Docket Y0997069 in US Rating: 2 Pts:3

Co-inventors: Neumayer, D.A.

Title: THIN-FILM FIELD-EFFECT TRANSISTOR WITH ORGANIC SEMICONDUCTOR REQUIRING LOW OPERATING VOLTAGES 09/11/96 Opened as Discl Y08960358 Status:Filed

Action: File 03/04/97 Discl Review

03/25/97 Filed as Docket Y0997057 in US Rating: 2 03/12/98 Filed as Docket Y0997057 in KO. Rating: 2 Rating: 2 Pts:3

04/10/98 Last Office Action

Co-inventors: Purushothaman, S. Dimitrakopoulos, C.D. Furman, B.K. Neumayer, D.A. Laibowitz, R.B.

X Title: HIGH DIELECTRIC CONSTANT, BARIUM LANTHANUM TITANATE THIN FILM CAPACITORS FOR RANDOM ACCESS

06/20/96 Opened as Disc1 Y08960255 in US Status:Opened Co-inventors: Gupta, A. Shaw, T.M. Laibowitz, R.B.

Title: METHOD FOR FORMING NOBLE METAL OXIDES AND STRUCTURES FORMED THEREOF Status:Filed 10/30/95 Opened as Discl Y08950450 11/12/96 Discl Review Action: File

Rating: 2 11/05/97 Filed as Docket Y0996239 in US 10/20/98 Filed as Docket Y0996239 in JA Rating: 2

07/30/98 Filed as Docket Y0996239 in TA Rating: 2

Co-inventors: Schrott, A.G. Saenger, K.L. Hummel, J.P. Neumayer, D.A. Laibowitz, R.B.

Title: PEROXIDE ETCHANT PROCESS FOR PEROVSKITE-TYPE OXIDES 10/23/95 Opened as Discl Y08950434 Status:Filed 08/08/97 Disc1 Review Action:File

(9) 04/08/98 Filed as Docket Y0997256 in US Pts:3 Rating: 2 Co-inventors: Rosenberg, R. Cooper, E.I. Laibowitz, R.B.

Title: RF TRANSPONDER FOR METALLIC SURFACES 08/02/95 Opened as Discl Y08950329 in US Status:Opened Co-inventors: Afzali-ardakani, A. Feild, C.A. Duan, D.W. Brady, M.J. Moskowitz, P.A.

Title: METHOD FOR CLEANING THE SURFACE OF A DIELETRIC

09/06/95 Opened as Discl F18950292 Status:Filed

09/06/95 Sent to Evaluator

02/05/96 Evaluated Action: Search

04/19/96 Discl Review Action: File

12/06/96 Filed as Docket FI996047 in US Rating: 2 Pts:3

Rating: 2 11/29/97 Filed as Docket FI996047 in KO 05/26/97 Filed as Docket FI996047 in TA Rating: 2

06/11/98 Last Office Action

Co-inventors: Kotecki, D.E. Wildman, H.S. Yu, C. Natzle, W. Laibowitz, R.B.

Title: NANO PHASE FABRICATION OF COPPER-GLASS CERAMIC COMPOSITE VIAS IN CORDIERITE SUBSTRATES

10/05/92 Opened as Discl Y08920907 in US Status: Published

10/08/92 Sent to Evaluator

Action: Publish 12/17/92 Discl Review

01/06/93 Mailed to Tech Discl Bulletin 09/02/93 Published Pts:1

Co-inventors: Kang, S.K. Shaw, T.M. Brady, M.J.

Title: METHOD OF SINTERING ALUMINUM NITRODE

Status:Closed 11/06/92 Opened as Disc1 FI8920668 in US

11/06/92 Sent to Evaluator

12/18/92 Closed

Co-inventors: Takamori, T. Shinde, S.L.

Title: METHOD OF SINTERING ALUMINUM NITRIDE

Status:Closed

11/06/92 Opened as Discl FI8920667 in US

11/06/92 Sent to Evaluator

12/18/92 Closed

Co-inventors: Takamori, T. Shinde, S.L.

Title: ALUMINUM NITRIDE BODY AND METHOD FOR FORMING SAID BODY UTILIZING A VITREOUS

SINTERING ADDITIVE

08/13/92 Opened as Discl FI8920525

Status:Filed

08/17/92 Sent to Evaluator

09/29/92 Evaluated

Action: Search

12/23/92 Discl Review

Action:File

05/10/95 Filed as Docket FI992168B in US

Pts:3 Rating: 2

05/28/96 Issued as Patent 5520878 in US

Co-inventors: Takamori, T. Shinde, S.L.

Title: ALUMINUM NITRIDE BODY AND METHOD FOR FORMING SAID BODY UTILIZING A VITREOUS

SINTERING ADDITIVE

08/13/92 Opened as Discl FI8920525

Status:Filed

08/17/92 Sent to Evaluator

09/29/92 Evaluated

Action: Search

12/23/92 Discl Review

Action: File

12/22/93 Filed as Docket FI992168A in US

Rating: 2

Pts:3

Pts:3

01/09/96 Issued as Patent 5482903 in US Co-inventors: Takamori, T. Shinde, S.L.

Title: GOLD DOPING OF YBA2CU307-8 AS A MEANS OF INCREASING TRANSPORT CRITICAL

CURRENT DENSITY

02/12/92 Opened as Discl Y08920161 in US

Status:Closed

02/14/92 Sent to Evaluator

05/15/92 Closed

Co-inventors: Daeumling, M. Shaw, T.M.

Title: PROCESS FOR PRODUCING CERAMIC CIRCUIT STRUCTURES HAVING CONDUCTIVE VIAS Status:Filed

07/19/89 Opened as Discl Y08890552

07/25/89 Sent to Evaluator

08/10/89 Evaluated

Action:Search

07/30/90 Discl Review

Action:File

12/17/92 Filed as Docket Y0990091B in US

Rating: 2

08/16/94 Issued as Patent 5337475 in US

Co-inventors: Vallabhaneni, R.V. Giess, E.A. Farooq, S. Cooper, E.I. Kim, Y.H.

Vanhise, J.A. Aoude, F.Y. Muller-landau, F. Shaw, R.R. Walker, G.F. Rita, R.A.

Neisser, M.O. Park, J.M. Shaw, T.M. Brownlow, J.M. Kim, J. Knickerbocker, S.H.

Title: VIA PASTE COMPOSITIONS AND USE THEREOF TO FORM CONDUCTIVE VIAS IN CIRCUITIZED

CERAMIC SUBSTRATES

07/19/89 Opened as Discl Y08890552

Status:Filed

07/25/89 Sent to Evaluator

08/10/89 Evaluated

Action: Search

Action:File

07/30/90 Discl Review

03/20/91 Filed as Docket Y0990091A in US

Rating: 2 Pts:3

02/01/94 Issued as Patent 5283104 in US

Co-inventors: Vallabhaneni, R.V. Giess, E.A. Farooq, S. Cooper, E.I. Kim, Y.H.

Vanhise, J.A. Aoude, F.Y. Muller-landau, F. Shaw, R.R. Walker, G.F. Rita, R.A.

Neisser, M.O. Park, J.M. Shaw, T.M. Brownlow, J.M. Kim, J. Knickerbocker, S.H.

Call your award coordinator, IPL department, or T/L 826-2680 for help.



#### CONTRIBUTION REVIEW SELF-INPUT

NAME: <u>Puncombe Peter R</u>

Emp. Ser: 155139

Date: 10/23/95

- T.R. McGuire, A. Gupta, P.R. Duncombe, M. Rupp, J.Z. Sun, R.B. Laibowitz, W.J. Gallagher & G. Xiao "Magnetoresistance and Magnetic Properties of (La<sub>1-x</sub>)MnO<sub>3-6</sub> Thin Films" 3M Conf. Proc: 4/96
- T.R. McGuire, P.R. Duncombe, G.Q. Gong, A. Gupta, X.W. Li & G. Xaio "Magnetoresistance & Magnetic Properties of (La<sub>1-x</sub>)MnO<sub>3-6</sub> (Vacancy) Bulk Materials" 11/96 3M conf CMR Open Forum entry
- J.Z. Sun, L. Krusin-Elbaum, A. Gupta, G. Xiao, P.R. Duncombe, W.J. Gallagher & S. P. Parkin "Magneto-Transport in Doped Manganate Perovkites" 3M conference 11/12-15/96 Atlanta, Georgia
- P. Lecoeur, A. Gupta, P.R. Duncombe, G. Gong & G. Xiao "Emission Studies of the Gas-Phase Oxidation of Mn during Pulsed Laser Deposition Managanates in O2 & N2O Atmospheres" JAP 80(1), 7/1/96
- J.Z. Sun, L. Krusin-Elbaum, A. Gupta, G. Xiao, P.R. Duncombe, W.J. Gallagher & S.S.P. Parkin "Colossal Magnetoresistance in Doped Manganate Perovskites" IBM J&D to appear 1996/97
- A. Gupta, G.Q. Gong, G. Xiao, P.R. Duncombe, P. Trouilloud, P. Lecoeur, Y.Y. Wang, V.P. Dravid, & J.Z. Sun "Grain Boundary Effects on the Magnetoresistance Properties of Perovskite Manganite Films"
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- P.R. Duncombe, S.L. Shinde, & T. Takamori "Aluminum Nitride Body & Method for Forming Said Body Utilizing a Vitreous Sintering Additive" US05520878 issued 5/28/96; I.A. Patent issue Award: 8/96
- Ali Afzali-Ardakani,: Mike Brady, Dah-Weih Duan, Peter Duncombe, Chris Feild, and Paul Moskowitz "RF Transponder for Metallic Surfaces" Docket#:YO895-0329 submitted: 8/2/95
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- C.D. Dimitrakopoulos, P.R. Duncombe, B.K. Furman, R.B. Laibowitz, D. Neumayer, S. Purushothaman, J. Shaw
   "Field Effect Transistor for Low Voltage Operation" Disclosure YO896-0358 rated file: 9/11/96
- R.B. Laibowitz, P.R. Duncombe, D. Neumayer, K.L. Saenger, A.G. Schrott "Noble Metal Surfaces" YO896-04xx rated "file" 10/96
- T. Shaw, R.B. Laibowitz, P.R. Duncombe & A. Gupta "High Dielectric Constant Barium Lanthanum Titanate-Based DRAM Structures" Disclosure #: YO898-0681 rated File 5/96 in preparation
- D. Neumayer, P.R. Duncombe "Fabrication of Barium Strontium Titanate Films" YO896-04xx rated File 10/96 in preparation

IBM Commitments:

To Win

To Execute

To Teamwork

#### ATTACHMENT B

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Technical Notebook

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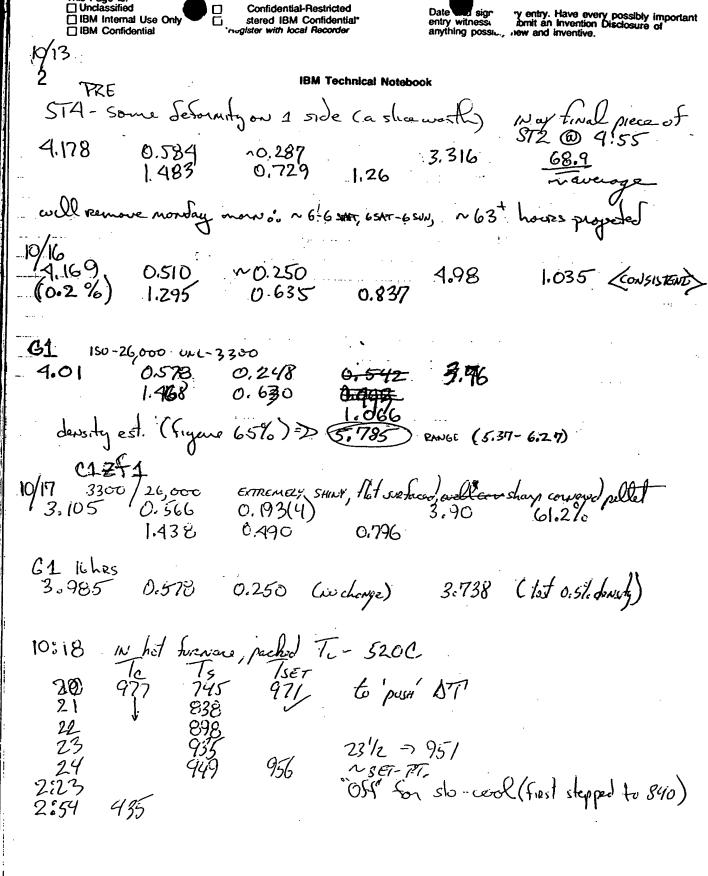
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Date of Last Entry

11/12/87

6/88

Security Classification:



10/17

# **IBM Technical Notebook**

C12+1 > pellet multiply anadred as if organic possible vaporized, evidence of rapor transport to species place, etc. ex Not

2.925 5.5%

81,00

9.79-3.105=> 6.685

10/18

GI - post 4.094 split in 4 pieces (seeminger on cooling)

GZ 1.1

0.579

0.253

1.093

pellet slightly distinct

33

4.155

0.510 0.220 1.295 0.559

0.736

5.64 about expeded density

DDI PRA

3,10 0.5765

0.191

30:4

0,513

0.165

0.559

5.61

38.2

entry. Have every possibly important mit an Invention Disclosure of and inventive.

**IBM Technical Notebook** 

11/24 Thermodyne Tube Lunace set-up specstheraple: dia. ~0.255 length 20"+ JUSTO 23

Ser-up complète of plug in jacks jecl wire, S cooples.

11/30 Analytical Sibmission

Your Burge Coo y Bay Cu

Sitils pre 2.0

C6post mice

DRC 123. **C7** 9:0

C8 DD 123

09 off comp 2:11

y Bu, a

y, Bu, Cu

bly important sure of



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☐ Registered IBM Confidential\*
\*Register with local Recorder

5

New 309 GRINDING CHARGE of SITIO3 in MILL (3:10)
Oz compressed AIR, CO2 cylindens obtained of regulations
An

VIELD -> 20.4g is MUST Be some from 20 batch or 2002 Combined my OLD purde -> 23g of miled powder

12/2
C1. both 156 geams left
39.56 g (ribg kept for files

10.529.5 left for pellets

10 for gurony change TFE/Toluane

New BOTTLES GEDERED, NO TEFUN AVAILABLE, - OPPROX- 60 hes total

St. T. O3 pellet 5 => 10-10/29 hes down NA: 2-2 m (N/2)/N/2-12 Thus 24)

STA, ST6 - start 10 AM. 12/18, numerous interruptions due to fueroce moltunations, out 12:00 p.M. 12/10 ST5 edge chips I side OK otherwise 21 150.

\*14.08 0.285 0.584 0.52

(\$01) 0.237 0.520 4.94 1.027 0.602 1.321 0.825

STG long chip during 150 presong IN H4 side, must do

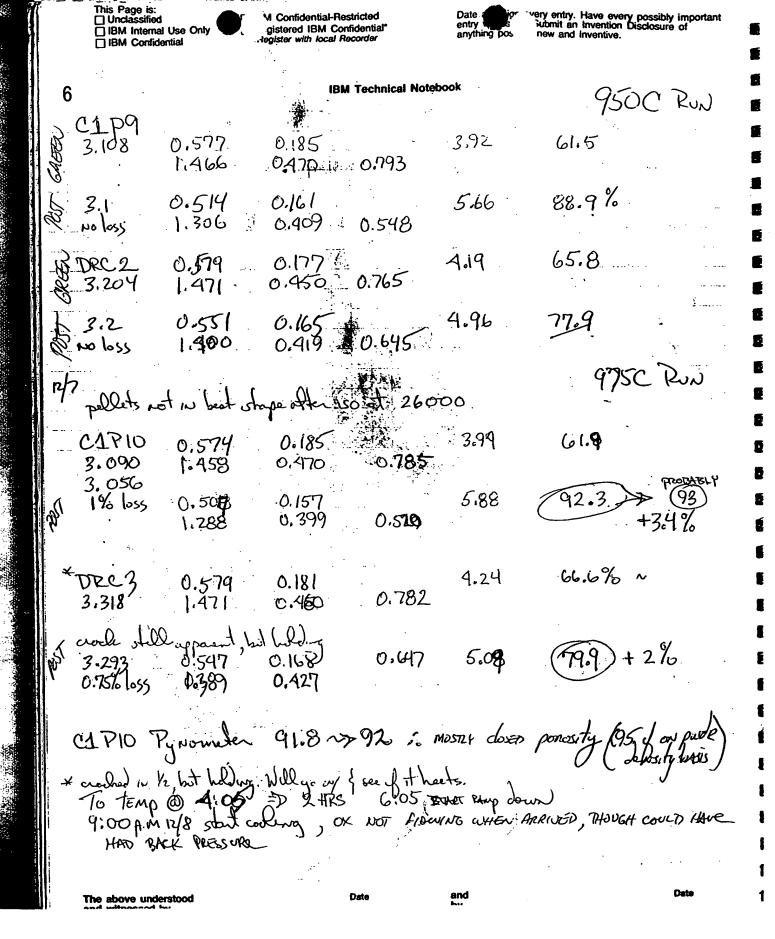
#4.128 0.686 0.286

4.15 0.513 0.249 4.92 1.023 1.303 0.632(5) 0.843

The above understood

Date

and



2.57

Rel. density calc 0.3 (6.00) + 0.7 (4.9) > 5.23 approx theoretical EMPIRICALD'S

2,825 0,90

The above understood

		e is: ssified nternal Use Only confidential	Confidential-Res jistered IBM Conf register with local Res	idential*	Date entry wheres anything possi-	ery entry. Have e Submit an Inventi y new and invention	every possibly important on Disclosure of re.
12	/9						
			IBM	Technical No	tebook		
		III. DENSITY MORKSHEET		III. DENSI	TY MORKSHEET		-
	,	BagCyO  ENHELE I.O. Fello Endo  SOURCE  TOTAL WRIGHT  TARE WEIGHT  LACT  ENHELE WEIGHT  LACT  ENHELE WEIGHT  LACT  ENHELE WEIGHT  LACT  ENHELE WEIGHT  LACT  ENHELE  ENHELE WEIGHT  LACT  ENHELE  ENHELE		TANKE WEIGH	SUMITEMO OPERATOR OF 12.606 PRE OUTGASSE	<u>0.9-87</u> *	५५ ३.८७ <u>१</u> .७०
		OPERATIONAL EQUATION $V_p$ $V_p$ = Volume of Founder (or $V_q$ = Volume of Sample Cei	= v <sub>c</sub> + \begin{bmatrix} V_A \\ 1 - \theta_2/\theta_3 \end{bmatrix}	OPERATION	L EQUATION $V_p = V_C + \begin{cases} V \\ 1 \end{cases}$ se of Powder (cc) so of Sample Cell Rolder (cc)	18.501 E	\$-464 \$-464
	٠.	VA - Added Volume P2 - Pressure Beading aft P3 - Pressure Sending aft R- 3.647 BOH 1	car Presserizing Coll ter Added V <sub>A</sub>	V <sub>A</sub> = Added F <sub>2</sub> = Press F <sub>3</sub> = Press		D 4.05 (+	5.079 2.394 2.51) 4.09 (4.55)
		*: 18.36.2 *: 4.980	18.488		18.520 18.562 (.078 (.213	18.539 18.539 5,222	· · · · · · · · · · · · · · · · · · ·
		\$ 0.24 (51) \$ 0.24 (51) \$ 155+5.13 }	4.88 e/ce e/ce	OCUSETY _	4.15 *** 396 390	Row Or	
			best quess - 5.9 sternifftsometer true powder demsity			FOMDER DENSITY	
	:	SAMPLE V.D. 211 SOURCE TOTAL METGET 1/6/2 TABLE METGET 1/6/01 SAMPLE METGET 1/6/01	DATE 12.4.87  OPERATOR PRO  1. OUTCASSING CONDITIONS  1. ADDED VOLUME: V <sub>h</sub> CC  CELL BOLDER VOLUME: V <sub>c</sub> CC	20.417) 4 061 16416	SAMPLE I.O. 123 SOURCE DEC SOURCE 10,026 9.  TABLE WEIGHT 4,062 9.  SAMPLE WEIGHT 16,963 9.  OPERATIONAL EQUATION V V.	ONTE 12-9-87 OPERATOR PRO- OUTGASSING CONDITIONS  ADDED VOLUME , V, 24,85 CELL BOLDER VOLUME, V, 26,6	Ω <u>ee</u> .
		OPERATIONAL EQUATION $V_p$ . $V_p$ = Volume of Powder (cc. $V_o$ = Volume of Sample Cel. $V_A$ = Added Volume $V_A$ = Added Volume $V_A$ = Pressure Reading aft	1 Holder (cc)		Vp = Volume of Powder (cc) Vp = Volume of Sample Ccll No Vp = Added Volume Pp = Pressure Beading after Pr Tp = Pressure Beading after A	ressurising Coll	· .
	• •	P <sub>3</sub> - Pressure Reading att  R- 165  R- 165  R- 18.557  P <sub>3</sub>	R-3.663 R-3.663 R-3.663 R-3.663 RDH 7  RDH 7	8.529 5.098	*, <u>{018</u>	PATA // R: 3.4428 EDH 3 EDH 2 EDH 3 EDH 3	· 
		v, 2.578 cc  DESSITY 6.05 e/cc  \$ 0.3  (\$95-6.35)	cc 2.036.cc 9/cc 6.0000-/cc	2.748 5.97	V, <u>2.13 cc</u> DESSITY <u>6.21 s/cc</u>	9/cg	
		total 2605		1.4.			

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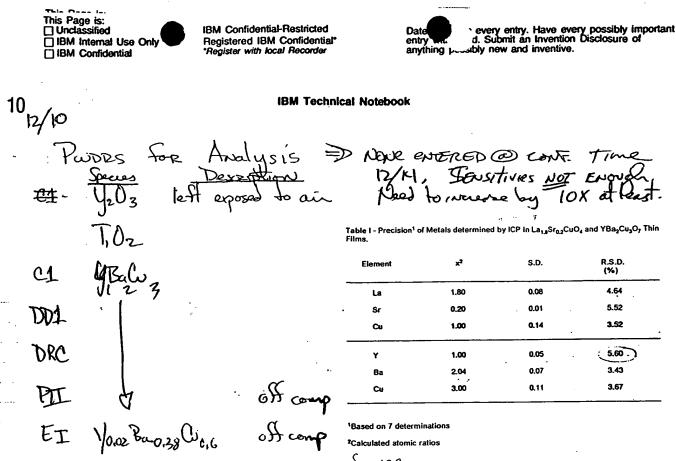
## **IBM Technical Notebook**

	. IDM TOCHNIC	281 11010D0011
•	III. DENSITY MORKSHEET	. III. QUASTIY MORKSHEET
	STEREOPYCHOHETES TRUE FOMDER DEMSITY	STEREOPYCHONETER DAN 2. TRUE PONDER DEMSITY
	SOURCE DIMANS OPERATOR (TC)  TOTAL MEIGHT 19,700 9. OUTGASSING COMPUTIONS  TAKE MEIGHT 4,061 9. NOOED WOLUME, V. BCS2 cs  CELL MOLDER VOLUME, V. 31.85 cc	EAMPLE II.D. 123  BOUNCE CL. OFFEATOR PRD  O
	OPERATIONAL EQUATION $v_p = v_c + \begin{bmatrix} v_A \\ 1 - e_2/e_1 \end{bmatrix}$	OPERATIONAL EXCIPTION $v_p = v_c + \begin{bmatrix} v_A \\ 1 - v_2/v_3 \end{bmatrix}$
	$Y_p$ = Volume of Powder (cc) $Y_c$ = Volume of Sample Cell Bolder (cc) $Y_a$ = Added Volume $Y_c$ = Pressure Beeding after Pressurising Cell $Y_c$ = Pressure Beeding after Added $Y_A$	$\Psi_p$ = walums of Powder (cc) $\Psi_c$ = volume of Sample Cell Holder (cc) $\Psi_A$ = Added Volume $P_2$ = Pressure Reading after Pressurizing Cell $P_3$ = Pressure Reading after Added $\Psi_A$
	R=3.646 R>3.646 R	E+ 5.68. 809.1 809.2 809.2
	18.63 18.561 18.561	· <u>18.508</u>
	5.103 5.091 5052	·, <u> </u>
	v, <u>2.523</u> cccc	y, <u>3.082 cc</u> <u>cc</u> <u>cc</u>
	DESITY 6.199 a/cc a/cc	DESSITY 6,13 m/cc a/cc a/cc
	no broth	•
	STREEDPTCNOVERER  THUE POWDER DENSITY  SAMPLE I.D. 123 DATE 12-9-87  SOURCE CA-2-cen OPERATOR PRODUCT OF THE TOTAL METIOR I (4.59 a. OUTCASSING COMDITIONS  TARE WEIGHT 10.30 a. ADDED VOLUME . V. 84.57. SE  CHIL MOLDER VOLUME . V. 84.57. SE  CELL MOLDER COMMITTED  V. 1 - Prussure Reading after Pressurizing Call  F. 1 - Prussure Reading after Added V.  R. 3 - SB  R. 1 - Prussure Reading after Added V.  R. 3 - PRUSSURE Reading after Added V.  R. 3 - PRUSSURE Reading after Added V.  R. 3 - SB  RUH 1 - RUH 1  F. 18.679 18.641  V. 1.703 CE  CEL MOLDER COMMITTED  CE	SIREDPYCHONETER  THUE FORDER DERSITY  SOURCE CA1-975287 OFFEATOR STED  WITCH MEIGHT 4.612 9. OUTCASSING CONDUCTIONS  THAN MEIGHT 4.61 9.  EMPLE MEIGHT 4.552 0. ADDED VOILING V. M.
	16	- 1875

The above understood and witnessed by

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and by \_\_\_



- John 153

Theoretical cight % cales. 18	BM Technical Not	tebook	11	
T.O2 > 47.90/19.8988 >> 5		And 1 57.3	CREAR Reported.	
Src03 => 87.62/147.62935->	59.35		PCT ANALYZETD	
Bally => 137.34/197,34935 ->	69.59@			
<i>I</i>	89.566	88.9	99.26	
Sition Sr > 47.74(5) Tr > 26.10(1)	M.W. 183.518	2		•
16 = 26.10(1)			T 2-2-9 11-9 1	

**C6** 

85.05% (15% pool) "3.48% RID" C5-92.72 (7.3% poor)

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					Thom ISHAW
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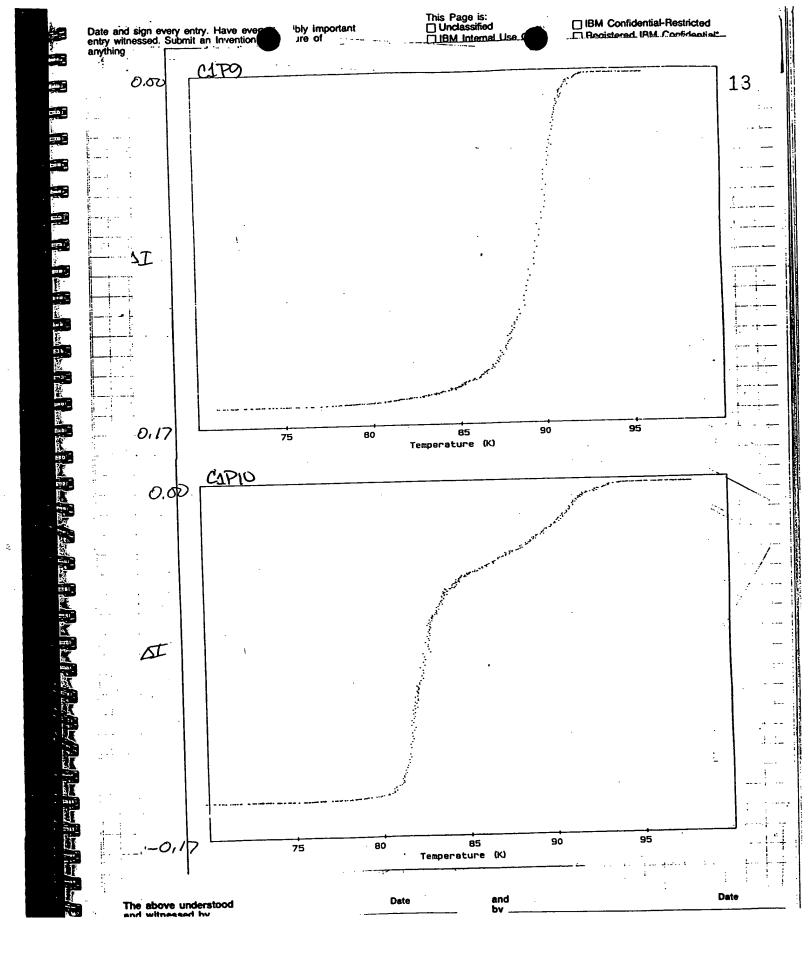
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IBM Technical Notebook

Except on 1 bump (0.06 5 veovered) => very smooth, overestful preparation. Placed in drying over for weekend drying. (over cleaned before use also)

12/21 after breaking up colse and re-balang under vac @ 70°C for 3 thes.

Brisk#1 transferal ideally wort >5 percux

tare \_ 86.21 80.26 greeney

97.74 68.29 226.49 expected 227.46 (99.57%) 0.3 gr recovered on burding blee,

CRX2 172.72

941.98

77.74

+.03 "moony"

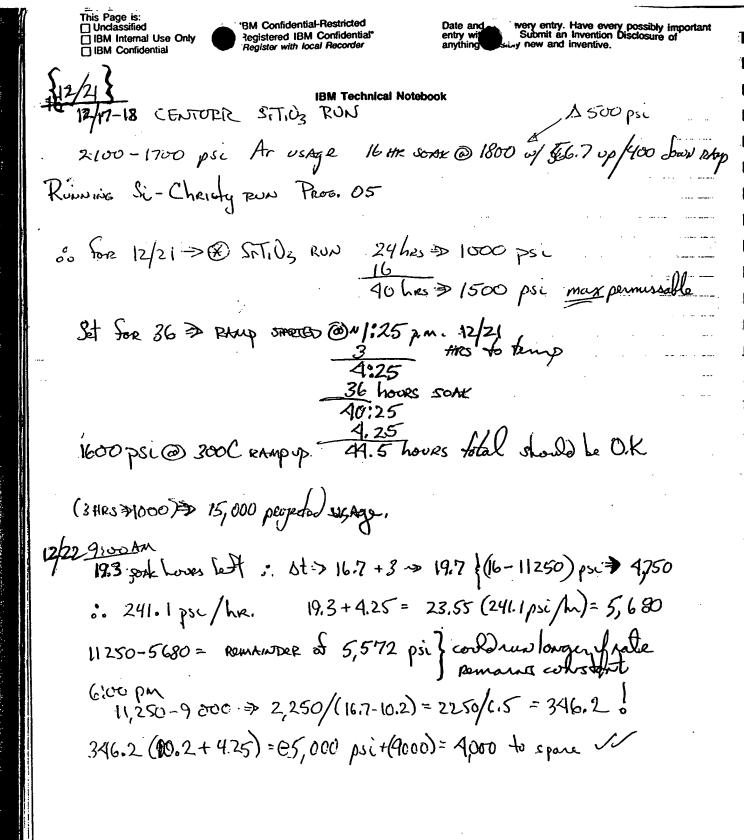
226.79 LdQ 99.7%

227616

CEX 3 173.46 105.16 "Let" 68.29

RXW. Run 1 => W & onto All Oph 320 Ramp to 940C, 450 cool Ramp 14 hres + 3 up + 2 down = 20 hrs total

12/22 1170 1 2 112 1 20 120



# IBM Technical Notebook

12-22

C2 RXN SeeMS GOOD, NO APPARENT LIQUID, LARGE SHRINKAGE NO VISIBLE GREEN, GOOD BLACK COLOR, BEFORE UNLO ADING.

CRUX #1.

17 BARCED, BURGHAT.

<u>6.21</u> *b*1 2.76

80.76+(80.76×(-0,1182))= 71.214

loss carc.

227.46 Reveted purder

120:54 = 0.52994 wft % & CO3

crux<sup>#</sup>z wital 172 94

172.72 <u>94.98</u> 77.74

valore = 68.55

153.34 (0.52994) = 0.41176

D= 0.52994-0A(176= 0.1182% total

crus#3

173.46
105.17

as above

= 60.218 199.983 total

= 200.58 UV OK.

Actual Viels - 19 HR PXN @ 9400

ceux #2 |

170.45 **94.9.8 34.24**  total wift isotal tare

ABOOK ELVECTED & EXIS

2.279 85 89.44

75:47

6.919 (**9.589)** 

Crex#1

169.92 86.21 78.71 78.

7,496

(9,546)

21.5

24.7

cex#3

171.91

1.55g oght 105.17 66.74

6.522

19.2

(8.015)

21.8% O.K

The above understood

and over.



18 Reclack of with loss cole.

**IBM Technical Notebook** 

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153,34 = 0.776995 (120,54) = 93.6589999 126.881 g

26.881 g /3 cmc = 2 8.96 g/cruelle 2 correct

Individual right inoxieres diving grending

12/29

Oux 3 66.72 whooled

171.82 looder 171.91 prevensly -09 0 loss = 0.135%

169.38 171.82 -2.44 bs> + -1.55 3.99

CRUX (2) 86.20 tare (6.19/20)
78.75 load (-0.02)78.73
78.69 gam often grunding
164.85
78.65 0.09g loss = 0.015/6 los

161.68 164.85 -3.17 bss + -2.27 5.99 bbl bodate

CROX (1) 75.49/8 UN loaded 941.99/8 tare 100d (pecux)

167.18 170.44 - 3.26 loss 2.05 5.31 

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Was 8	25% reades los so da	before the	ers thou	2432	g/27.46	(88.5-85.2)
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Look	la 283	g total.	کون نهمون			· · · · · · · · · · · · · · · · · · ·
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	COMPS SCRIPT A1 dated 87/12/02 14:32:	25 Page 1	7	· · · · · · · · · · · · · · · · · · ·
	•			R- mestical
	Date: 2 December 1987, 13:24:31 EST From: PLECHAT at YKTVHZ To: PRD		· •	1
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	# C1 Y Ba Cu O 0.03 0.68 X	Cu=1, ICP	·,	1-12 0 4177
	# C2 Y 78.1 % (W/W)		\_?r	consistenty hex dire
	# C3 Ba 88.9 % -	-		<del> </del>
	# C4 T1 57.3 % -	/ error due to static electr. during weighing of somple!/		
	# C5 Ti 22.2 Y - Sr 49.4 Y -			
	# C6 Ti 24.2 % - Sr 50.6 % -			
	# C7 Y Ba Cu O 0.34 0.71 X 4	Cu=1	<u> </u>	
	#C8 Y Ba CuO_	<u>.</u> •	; ,	
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	2.37 1.10 X		<u> </u>	
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**IBM Technical Notebook** 

with 197.59

196.56 1.03 q lost danny granding

Must be totally converted @ this point.

7,000/0.371 = 18,870 PSL

23

C2P2 > WD leave votes on publi oler too busy

DIMIN 4.2 un PSD 10-1 ~ flat dist.

2.53 un aver, much better behaved pellet

0.734

green 27 150 pressed

3.96

wed @ 600°C => 20/up to 800, 10/40975, 20° down

0.554

0.186

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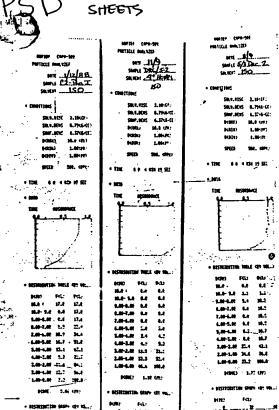
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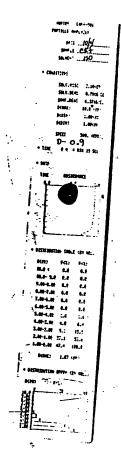
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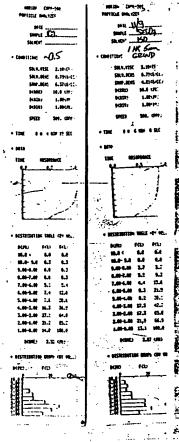
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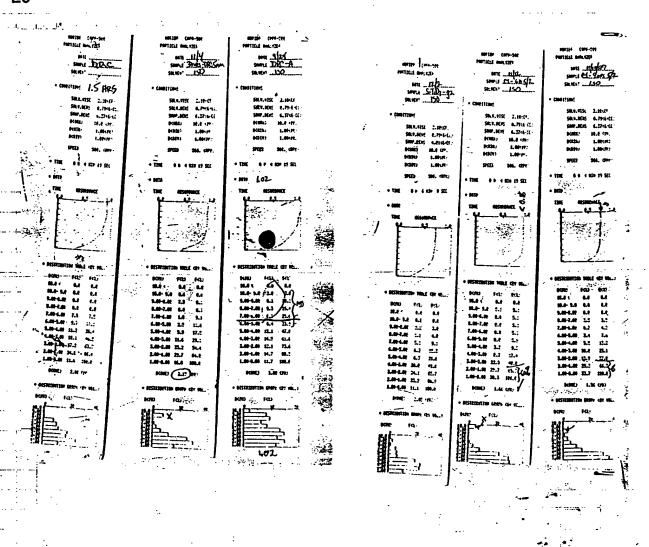
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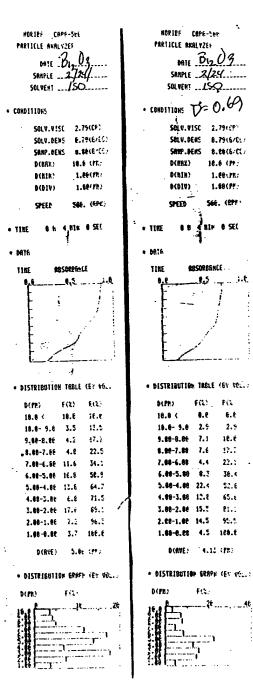
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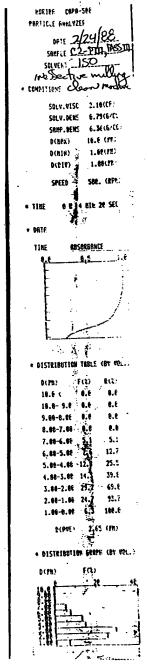
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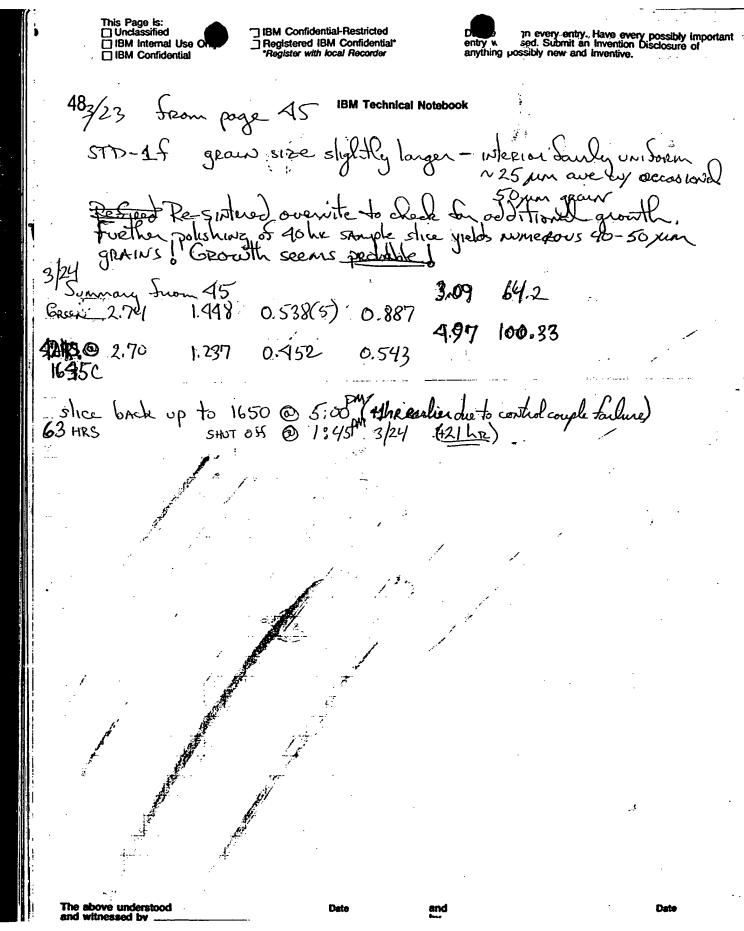
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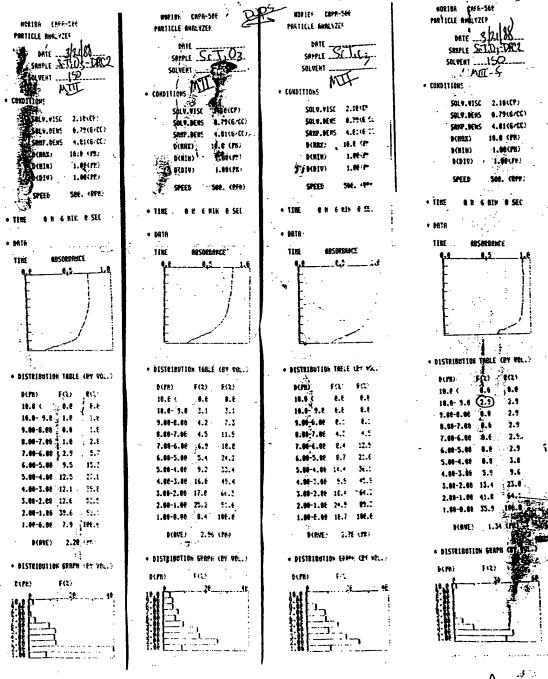
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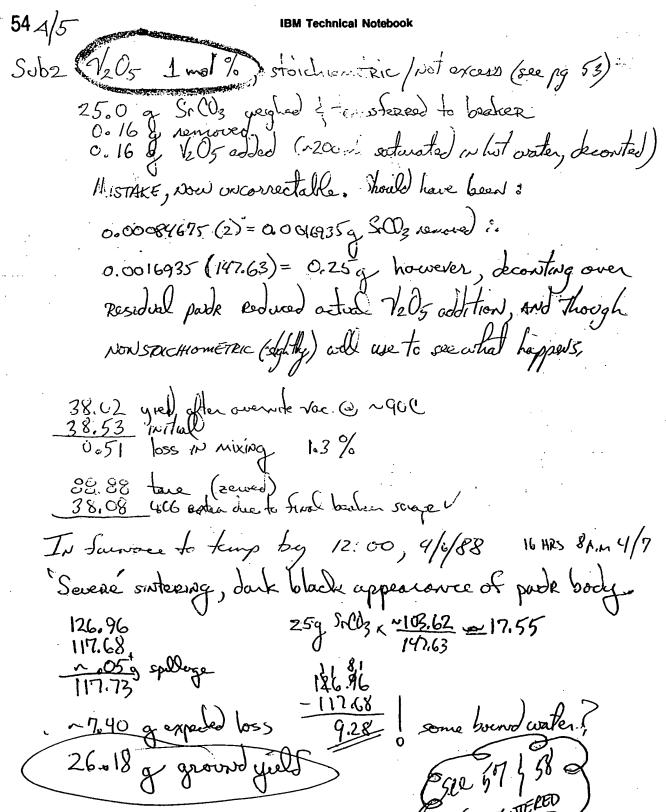
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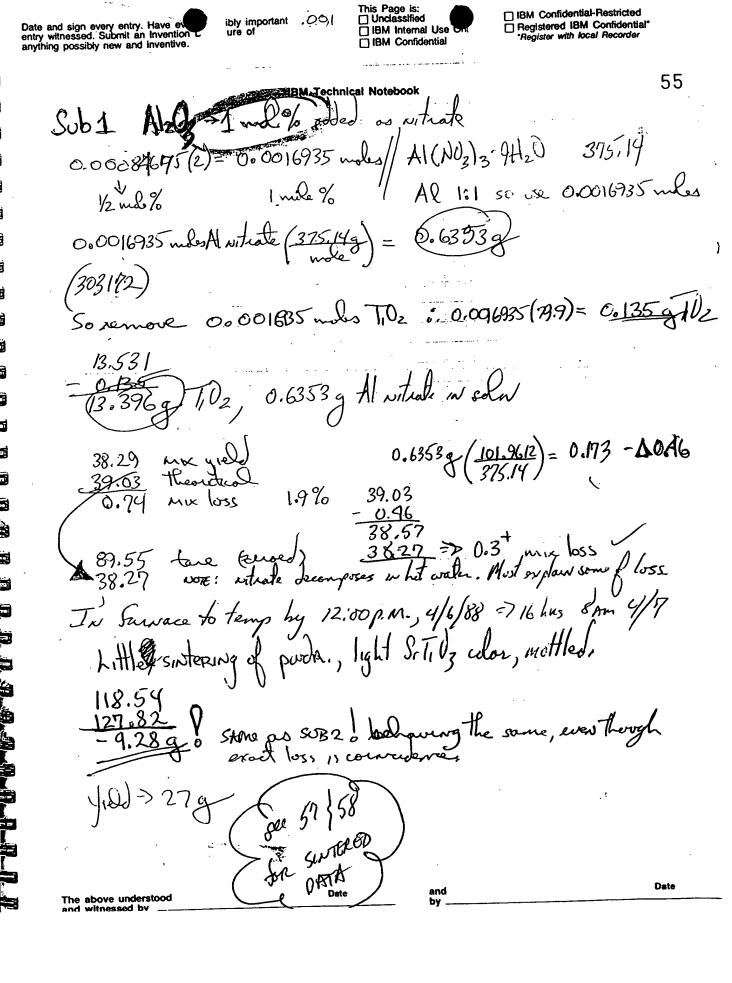
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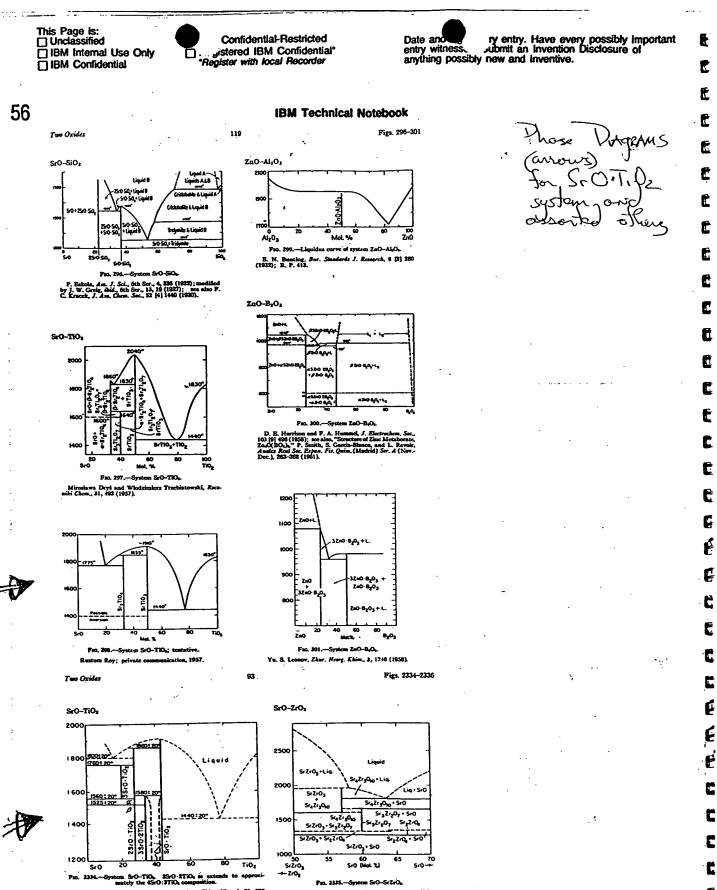
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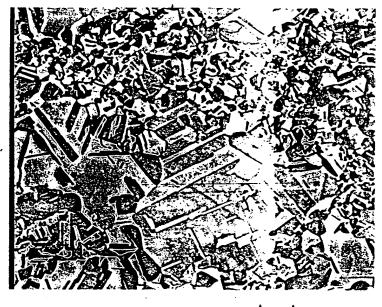
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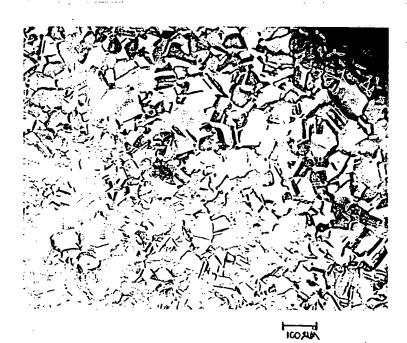
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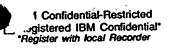
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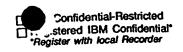
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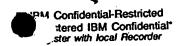
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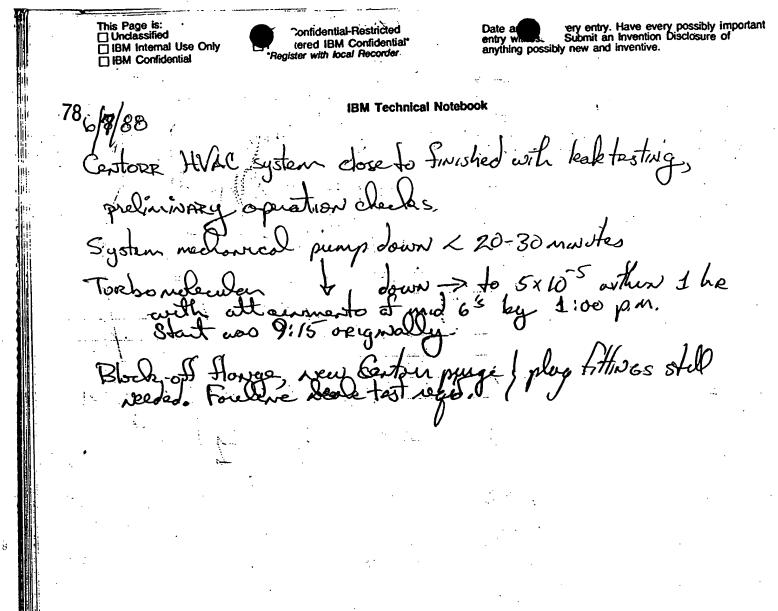
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	C3818	3.21	1.308	0.475 0.405	0.54 <b>%</b>	5.77-8	90.7-9		
	C3P/9	3.04	1.30 <b>6</b> 1.461	0.45%	•				
		2.97	1.298	0.386	0.511	5.81	91.35		
	C3P16	3.02	1.466	0,457					
	(1000)	• .			0.519	5.82	91.5	<i>\</i> 0:	
	C3P17	3.28	1.464	0.492	0.54	8 5.82	92.5	chip offe M	
===	•	(3.23)			· ·				
	The above and witnes	understood sed by	· ·	Date	<del></del>	and by		Date	



Date

The above understood and witnessed by \_\_\_\_

IBM	<b>Technical</b>	Notebook
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Bi/a Free-Cuable Sinter Vacuum Run

To glass shop 6/7/88. Batch sine to be 4.0 g to allow dark cose of manipulation during sealing of quarter tube.

4.0 (0.25) = 1.00g P

6 (0.75) = 3.00g Cu (will use 10 pm Cu parok)

Bi > 1.00 (0.99) Cu to <u>4.03</u> Cu 3.03 From JAR 4.02(1)

crux. w/mix 183 5.38(4) A.00(x) ~ 24.8 % ~ 25 75.2% 75

Repo - spiled in grass short

6/9 New crucible shape/size for the Lets take 5.0 g bother

1.25/5.0 (±0.01)

of my 8.35

6/14 Ofter overvite sinter & removal from avant tube

crue & sister 8.30 a (some spillage in the better

NO appreciable vapor peroduct stran loss

Conclusions: Vacuum doesn't appear to work as well as Ar/Hz. Sample Soll of holes, but no evidence of application, so holes are real. Again, no evidence of rapor phase deposition in take.

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806/14/88	IBM Technical N		Macatlon)
To do: 10,2 WENDS 6/22/88	0,25 % IN Ar/1	(2 (5) A 11 (	RUN
5 gram batches !	0.25 (5) = 1.25	8./3.75 Cu 2./ 4.00 Cu	
D 3 gran ny new con	ع الملمد		BAL ORDER
fore <u>1.03</u>	les small vo however la repeating, m	uds, some goodne ugwards a my s aybe longer time	gious versus(II)
1 9.00 true 4.00 5.00	seems very a majosopie ( or voids, V	sood, us large vo kom shows man	ry small pockets
6.31	ور احمد ال	sify fully	
- 5,00 Nomura	Q loss possible		
The above understood and witnessed by	Date	and by	Date

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رو المستقوم ومسا. 	MORIER CRPA-SEE	NORIBA CAFA-566 PARTICLE AVALYZEF - \	PARTICLE RNG_YZEF	
	RTICLE ANALYZER	6.19	DETE 3/2/188	
<u> </u>	DATE 3/24/00	SAMPLE SCT.D3-DACS	SAMPLE SETIOS-DACZ	
	SOUTH	SOLVENT15Q	SOLVEH"150	
	MI-MED	MII-S	+ CONCITIONS	
• 091	HOITIOHS WPROVEMENT	* CONDITIONS		
	SOLV.VISC 2.10(CF:	SOLV.VISC 2.10(CF. SOLV.DENS 0.79(E-CC)	SOLV.VISC 2.18(CF) SOLV.DENS 6.79(6/CC)	
	SOLU.BEHS 6.79(6/CC)	SRIP.DEHS 4.81(6/40)	SAME.DEMS 4.81(6-CC)	
<u> </u>	\$AMP.DEMS 1.81(6/CC) D(MAX) 16.6 (FM)	D(MRX) 18.8 (FR)	DINAX) 18.6 SPT;	
	D(818) 1.08(PE) -	D(HIH) 1.88(Ft)	D(H)H; 1.88(FF; P(D)H) 1.68(FF;	
<del></del>	D(D10) 1.88(PL)	\$(DIS) 1.88vf:		
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	)(FR) F(2) R(2)	0(PB) F(Z) R(Z) 10.0 (	18.6 4.6 8.6	
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	1.66-7.66 3.9 3.5	8.66-7.66 8.6 2.9 7.66-6.86 6.6 2.9	- 8.60-7.00 3.6 2.6 7.00-6.00 2.9 5.7	
	7.00-6.06 4.4 8.4	6.00-5.00 0.0 2.9	6.00-5.06 9.5 15.3	
	6.66-5.66 8.5 17.2 5.66-4.68 17.1 34.3	5.06-4.68 6.8 J.E	5.66-4.66 12.5 27.6	
	5.88-4.68 17.1 34.3 4.88-3.66 11.1 45.4	4.68-3.66 5.9 9.6	4.66-3.66 12.1 39.8 3.66-2.88 12.6 52.5	
.3	3.66-2.60 22.6 67.4	3.00-2.00 13.4 23.0 2.00-1.00 41.0 64.1	2.66-1.66 39.€ 42.1	
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MORIBA CAPA-Set	MORIBR CAPA-SEE PARTICLE ANALYZES	NORTH CAPE-SEE PARTICLE AMALYZEF	
PARTICLE BHRLYZEF  DRIE	DATE 5/19 SAMPLE C3-PI-T2 SOLVEN: 150	SMPLE C3-PT-T2. SOLVENT150	
• CONDITIONS	• CONDITIONS	• CONDITIONS  SOLV.WISC 2.10(CF:	
\$0LV.VISC 2.18(EF) \$0LV.DERS 0.79(6/CC)	SOLV.VISC 2.16(CF; SOLV.DEHS 6.79(E/CL)	SOLV.DENS 8.79(6/CC)	<u> </u>
\$889.0EKS 6.36(6/CC) 0(88X) 16.6 (FK) 0(81H) 1.86(FK)	SREPLOENS 6.36(6/CE) D(MRX) 10.E (17:) D(MIK) 1.88(17:)	DCD1A) 1106(141) — BCHIN) 1106(141) — BCHBX) 1016 (141, —	
(- 8(DIU) 1.86(Ph)	D(DIV) 1.68(Fh)	SPEED SAR. (RFH: -	
+ TIME 6 H 4 FIH 26 SEC	* TIME A H 4 KIN 20 SES		:
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TIME MESORRANCE	TIME MESORRANCE	• • • • • • • • • • • • • • • • • • •	
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10.0 9.6 9.6 9.8 10.0 9.6 9.6 9.8 9.00 8.80 3.4 13.2	10.0 ( 36.4 36.4 10.0- 5.6 3.2 35.6 9.00-0.06 4.3 43.5	16.6- 9.6' 2.5 29.6 9.66-6.66 4.1 33.7 8.66-7.66 1.5 35.2	
8.48-7.88 1.5 14.6 7.88-6.88 4.5 15.2	8.86-7.86 5.5 49.4 7.86-6.86 9.8 59.2	7.00-6.00 4.6 39.9 6.00-5.00 7.9 47.6	
5.00-5.00 11.2 30.4 5.00-4.00 14.1 44.5 4.00-3.00 15.6 60.1	6.68-5.66 E.3 67.5 5.68-4.66 E.: 75.6 4.64-3.66 9.5 E5.4	5.08-4.88 13.8 61.6 4.09-1.08 12.3 73.5 3.08-2.08 12.7 86.c	
3.60-2.66 19.4 79.5 2.64-1.66 18.6 97.5	3.06-2.06 6.2 91.6 2.08-1.06 7.8 99.3	2.68-1.86 12.2 76.5 1.68-8.86 1.2 186.8	
1.88-8.88 2.5 186.6 	1.00-0.00 0.7 100.t D(AVE) 6.94 (FE)	D(RVE) 4.84 (FR)	
- DISTRIBUTION GRAPH (BY VOL.)	• DISTRIBUTION GRAFK (BY SCL.)	• DISTRIBUTION GRAPH (BY VOL.)	
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		£:88 <u>1</u>	

Administrative Notes 3- RUSULTS: NECK MORIBE CAPA-586 MORIBA CAPA-SEE PORTICLE REALYZES PRETICLE REGLASES MATER MIE SIES RE SAMPLE C3-P2-T3 SAMPLE CO-STAMUX SOLVERT \_\_\_\_\_\_ SO\_\_\_\_ SOLVENT .... · CONDITIONS • CONDITIONS · CONDITIONS SOLV. VISC 2.18(CF) SOLV.RISC 2.10(CF) SOLU.DENS 8.79(6/CE) SOLV.VISC 2.10(CP) \$0EV.0ENS 0.79(6/CC) \$88P.DENS 6.36(6/CC) SOLV.DENS 0.79(6/CC) SAMP. DENS 6.36(6/CC) 18.8 (#1) SANP.DENS 6.36(G/CC) D(KAX) 18.8 (PK) D(MAX) 1.88(#%) DOMEST. D(MAX) 10:6 (Pt) 1.66(%) P(#163 1.00(7%) DOMEST ! 1.86(7%) 0(014) B(814) 1.88(PE: 9(017) 1.00(Fh) 566. (RPK) SPEEL SPEED 500. (RPE) SPEED 500. (RPR) -8 H. 4 HIN 26 SEC • TIME . TIME '8 H 4 MIN 28 SEC BH 4 MIN 28 SEC e TIME . DATE . . DATE D~ 0.9 . METR RBSERBRECE TIME > RESORERNCE TIME RESORBANCE TIRE . DISTRIBUTION THELE (BY VOL.) JABLE (BY VOL.) DISTE THE TABLE (BY VOL.) R(2) F(2) D(FR) R(2) €.6 e.e 18.6 ( B(FH) €.€ 1.8 16.4- 5.6 16.6 1.8 1.2 9.48-8.66 6.6 3.1 10.0- 9. 1.2 e.e 9.88-6.88 8.00-7.86 3.1 4.9 9.96-8.96 8.48-7.86 8.6 \$.\$6.7.88 2.4 9.2 7.00-6.08 4.3 1.7 7.88-6.8€ 8.€ 4.4 18.6 . 6.88-5.86 9.4 6.00-5.60 6.0 1.2 14.5 5.06-4.00 12.8 5.48-4.88 - 5.7 16.9 5.88-4.88 13.1 28.€ 4.68-3.66 26.7 52.2 4.46-3.66 27.7 38.6 4.00-3.08 6.5 34.5 69. 82.2 3.44-2.06 38.6 51.5 3.68-2.66 17.6 .2.00-1.06 14.3 96.5 67.4 100.8 2.06-1.00 1.00-0.06 3.5 188.6 106.6 1.00-0.00 D(UAE) 2"11 (LE) D(RVE) 2.63 (FE; D(RVE) 2.89 (FR • DISTRIBUTION GRAPH (BY VGL.) . DISTRIBUTION GOODH (EY VOL.) . DISTRIBUTION PH (RY VEL.: titt: FOO DCFR' F(k)D(FR)

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**Technical Notebook** 

Book I

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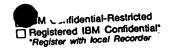
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12:57	1346	1350 :-	20,4	(700)	40	7.27	:	4.6 x/12 psi
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1:17	1550	1547	30.3	700	5.4	7.48	60.1	b 12_
1:48	_1.	1550	29.7	700	5.4	2.47	30	16/12
2:18	1550	1550	29,2	700 (90 <sup>†</sup>	5.25	7,469	0	125/3
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<sup>4</sup> 7/	5/88	S	TBX-	2	IBM Te	chnical Not	ebook 		·	
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3.07	~30%	<b>१९७</b> ० ।	0.8	∠300	۷۱ :	7.70		STOP S	166-	6
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517	200 VA 0	245	(O)	بب		7.61	dan da	Yacum.	troterso.	~125 PV/mw
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5130	8440)	134	GAS Prog o	ssf.	RALSO	e temp	, but A	r, GPM	ab 40 B	

Notes: 300 C@ 10:36: 2h 15m to S.P. => 12:45 (eit) + 2h => 2:45 Ryuptan N3.5 hes for woling, < 100 Copening @ 6:15 approx. (due to thermal mass lag)

Ousuccessful. Some stuckour to bottom foil. Cresked with diffusion zone and multiple phase boundary.

6/28, B./C.
Ar/Hz B.20

Bi 25 > 6

IBM Technical Notebook

Bi 25 pieces Remaining After slicing isostatically pressed to 28,000 PSI Of

Bi 25 > some obvious large your improvement on at least othe 1/2

By 20 -> possible visible evidence of compression, used to section.

Objectives:

- 1) isopiess as above
- 2) slice
- 3) onver Remaining sections

4)

study wetting /densification > temperature relationship

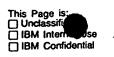
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			40.1					SE-5	RAMP 20/MW: 1200c/h Sma degramy > 1600
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12:55	2028	2003	84.4	J					Hold to continue Bout
1:04	2061 2083	H	<b>92.0</b> 96.0 98.0	···				48-4	Raised Momally PV steady & agreen
1:34	209/	H sam	<b>↓</b>	1000		· • •	•	3E-4 1AE-4 9E-5	Holding
2:30	2093 2093	و .	with	Dood	744	Acto		6E-5	Start PV RMup down
2:46	1727	ধ্য	1 38.9	790	7.5	_	-	25-6	Cool TAX for 1800C RUN
3120	1004	100	0 6.7	475	25	-	-	9.85-9	- · · · · · · · ·
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-4:38	)				m	anna	all	ζν ,	

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Pressures from 2/13/8		. \		
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Psy of 180 for Broth KSTAL INDUSTRATION	I Run 97III	resulted in yield	of moly stag an	1
a Western God	pprox. halve 1	kamb opion podiv	isig and apply	<b>.</b>
this des clust	Dones ST.		7III RUN YULDED O	Ly .
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10/14 I 88 Bxstal RIBM Technical Notebook

Sinal Proce of Juration Reak time Psig

OI 100°C O.1h Garr 110

O2 600h 2:8h 110
O3 1800C 475h - 4:75h 110 1.4-4.0 E-5 tork

O4 89h 21.2h Non 100e; when wife 60h know shipped

to 1730 int sortchouse baston 80/h

RAMP Kulkud IN. 4E-5-8E-7

Run motes! @ ~ 7:00 pm 7/3 TC leak noticed during inspection. See note above.

Results: Surely acrossed xstal Some sticking, but NO, RXW. with Mo shim on store, but controu Mo RXW from tops RAM/shim curple Pitting / Dungling top sentoce & likely initiating crocks.

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. . III. DENSITY WORKSHEET

## TRUE POWDER DENSITY

SAMPLE I.D		DATE 7/26/88
SOURCE 25	Be/Co Bar :	OPERATOR PRO
TOTAL WEIGHT	13.981 g.	OUTGASSING CONDITIONS
TARE WEIGHT	4.060 9.	
SAMPLE WEIGHT	9.921 9.	ADDED VOLUME , V 85.52 cc
•		CELL HOLDER VOLUME, V. 34.85 cc
		_

OPERATIONAL EQUATION 
$$V_p = V_c + \begin{bmatrix} V_A \\ 1 - P_2/P_3 \end{bmatrix}$$

 $\nabla_{\mathbf{p}}$  = Volume of Powder (cc)

 $\nabla_{\mathbf{C}} = \mathbf{Volume}$  of Sample Cell Holder (cc)

VA = Added Volume

P<sub>2</sub> = Pressure Reading after Pressurizing Cell

 $P_3$  = Pressure Reading after Added  $V_A$ 

P./P3 - 3,5415 3.5419 DATA

	RUN 1	RUN 2	RUN 3
P <sub>2</sub>	19.457	19.656	19.746
P <sub>3</sub>	5.494	<u>5.548</u>	5.575
v <sub>p</sub>	1.2006 ee	1.219 cc	1.206 cc
DENSI	ry 8,2634 g/cc	8,13 1ba/cc	<u>g/cc</u>
8.2	26/9.17 = 90% .	8.14/9.17 = 89%	8.23/9.17 = 89.75

Accessage: 89+89:75 ≈ 89.5 Between 69-90%

Conditions: 4000 m Nz

Conditions: 7500 W NZ

13

ENTERED 8/1/88

suitial compositions: & Bi (las) & Ci(ali)

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.... 50 \_.... (3)?

Lia. & B./Cu System Devlopment Sumary

NOTES: CONDITIONS AND & comp. Bi du sunt lead to distribute (surfacing) of plugs. No ocht loss. 5g somples av rilesous continu chicles

NOTES: crething or No does not soon to be tell possibly cride.
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The person's apprount. 10% too little he. No approvable word boss.

## **IBM Technical Notebook**

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D. Johnson	Sandres of 1ster	Lide)? Conditions: 25	mjs.
. 23	4	pund (77 %) from welling, syporting of howevery don't house much don'to hopped anninghyspi myskon swint og	•
Flor Eyr	A PE TA	tropp) and physic projection evident by	egt loss

Ar/Hz. No composition  Ar/Hz. No composition		E. 8.8. BC Carling: pelos pude auchle 25 belone Fue grafu \$ 7500 organis
Cir though 10 pm source to give better orweall Rosetts,  Rent parties Ren source to now more time, but cetting  characteristics source bother CVII never struck as went	t.	W Ar/Hz. No compostran
Cir though 10 pm source to give better orweall Rosetts,  Rent parties Ren source to now more time, but cetting  characteristics source bother CVII never struck as went	J	NOTES: Prosity is produced 30 orde has begindly with
Cir though 10 pm source to give better orweall Rosetts,  Rent parties Ren source to now more time, but cetting  characteristics source bother CVII never struck as went		as forming to welling soons capper complete little
characteristics spen better Cill recent sample at again	•	a though 10 pm soms to give better overall souths,
Ku (. sxounde)		characteristics spen better Cill recent stops at agot
	-	eus (; exovernte)

C. though 10 pm some to give better orace.  Rinetantion RIN some to now more the characteristics span batter Will receive strape con (: 2x000mm)	ll rosults, but wetting
F. Vacuum 25 Bi Row	
confusions: heally press, torque don't	ypen to asst

Joseph Toke. No endure of roper pla

Laster: 'plat' put cuelle see . salad @ 3500 oonwise as hofte. No competien: Realts: Cood avers and acting a both 20 \ 25 % samples, however 20% seems both Dogall with small con some pour chances 25% has some voids as well as pours, 20 \ 25% B. supho present to 27,500 psi postatically.
Some adapte of large powerty vaids. Little et 21 of Awaday esect: 0.02 | 0.04 of Bi-out ha.
From out of handsmituse you woulding. Powerly drawded.
Very good boking valually dure 20% shape. Boa: 25% Bi (100 hill) in priseous bout fast placed in house roccum (desseated) their sustand overwise per stot teachest producing recy good boding man-structure with little proserty.

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14

**IBM Technical Notebook** 

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Date

and

8-11-88

1500C

IBM Technical Notebook

15

GREEN phase substrate work

have one remaining substrate, Não-90% deuse, single phase, sinterT

1 pressed 0.2", 0.20 pellet et entertie

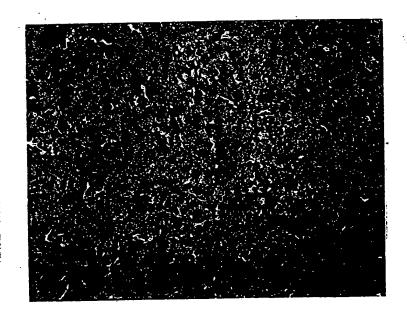
8-18-88 week sumary

pellet almost totally melts (2 d) with interaction between Alzig And 110. D.

pellet retrains \$15 INTEGRITY, But large Amount of LIQ forms 20, interaction of lia of and support 1400 C

lia & still present, though diminished. less interaction, for short sinter time 291 milled "on"

ulled purole. 13**45**C



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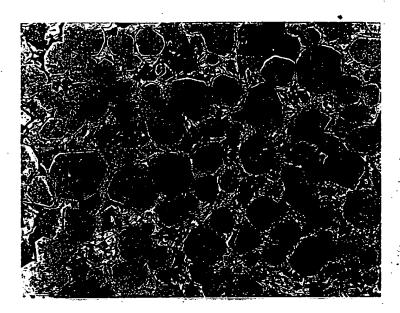
IBM Technical Notebook

1292C

18HRS

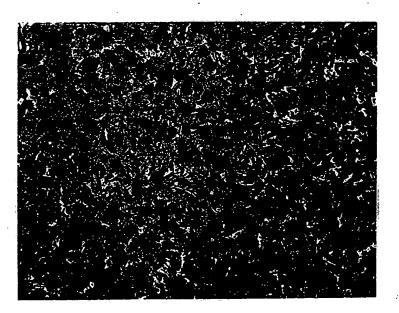
211 milled

(OOK



12650 'coarse' off comp' oversite

100X



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17

IBM Technical Notebook

211 milled 1235C

2HPS 100

1000 X



Condusion: sintering @ 1292C or higher capates 2 & material exagginated his & grain growth after prolonged period sintering @ 1235C closs not movice adequate sontering. pullet hemains green as apposed to higher temp where pellet trens block (presumably this is not simily service effect, but has chanced basis)

sintering @ 1266C may be optimal.

Purch delimitely ow?

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Date

end bv \_\_\_

#### **IBM Technical Notebook**

19

8-31

0.04g 0.83mm dia pellet set on edge of polished 211 substrate which itself pests on a piece of 211 pesting in a AlzOg boat on a bad of 123. Adjacent to substrate is small pellet of 211 to allow extent a pellet of 211 to allow extent is pellet to stradbe edge of substrate to minimize countries.

Heat teedment: 10°C/min to 1000C in Flowing 02 previous exps. in air/02 shound warrigevent melting of extetic @ ~ 1000C.

10:45 AM. To 5000 : 10000 plateau should be readed @11:35

Will allow to nett for 1h >> 12:35

10C/min >600 1:30 had

5C/min >600 1:30 had

> 0C/min -> 3000 quench

Flow not pronounced. Not alst of lia. formation. Pix taken.

Rado in Air/Oz where prev. exp. showed alot of ha. Sormation.

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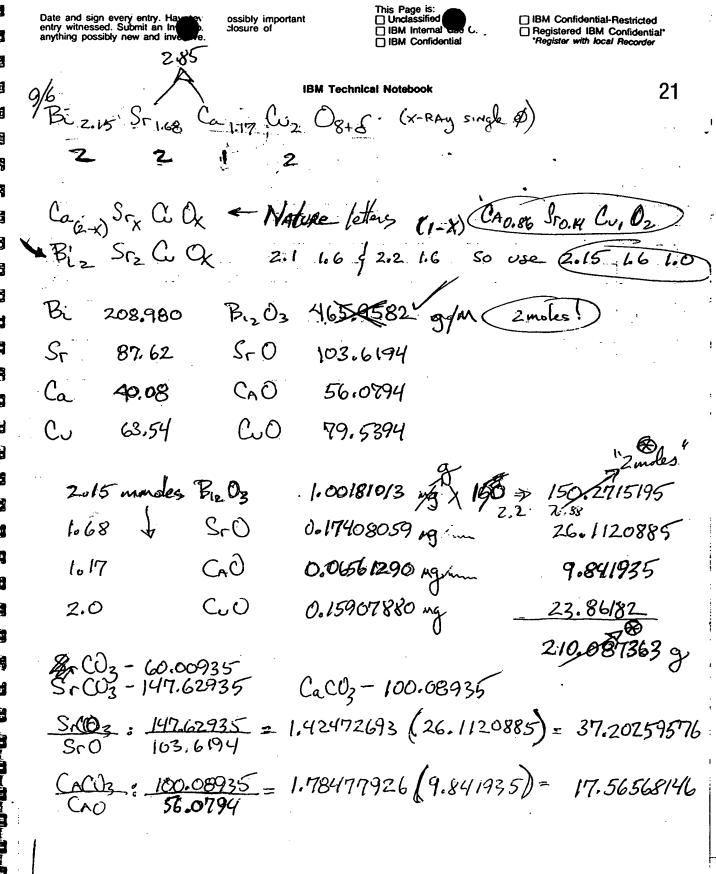
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Mixe) ground And columned @ 775 C 21 h (PE)	
3 gd and recolemned @ 850C for 16h	
(4) gra & null for pellet astron (intro) 2 molar correction!	
37.20259576 9.30064894 (0.70188889)= 6.528	
17.56568146 4.39142037 (0.56029328)= 2.4605  23.86182 5-965455  57.22540 - (0.7018889)-	
- 1.931 - 2.7776 52.522 g best Q	
28.176 13.950 (7075)=9.7895 4.16g bss	
13.950 (13.95 (.7075) = 9.7895 4.16 g loss 6.587 (0.560) = 3.69 2.9 g loss 7.06 total appears 57.661 (less COz loss*)	nnat
	nmn

:3

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ssibly important

249/9

**IBM Technical Notebook** 

BiSrCa Colonation II: gold lived Alz Os boot

86.51 bouly fite in larger boot

51.38 NS 51.43 0.1 % transfer loss

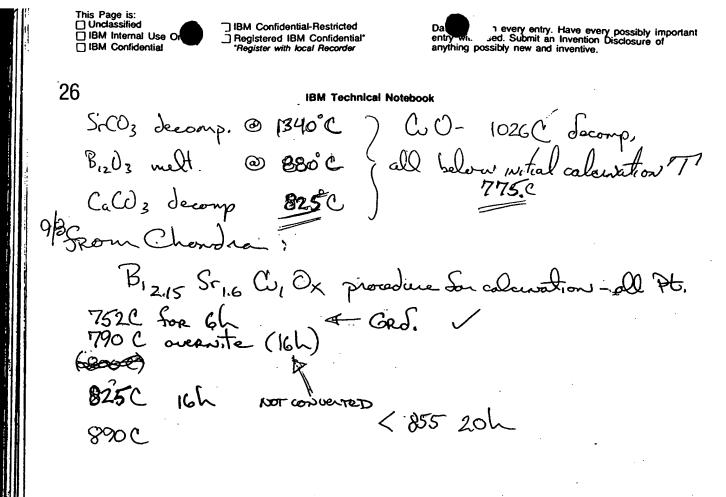
85.10 35.13 49.97

49.34 In fuerace (tube) for 850C, 16h coloniations

84.50(48) 24 stole procen

83.58 post 8500 16h columntin 35.13 post on X-RAY

25 **IBM Technical Notebook** Srue Co, Ox (ref. data pg 21) 2.15 mmoles B1203 1.00181013 but 2Bi = 1BiO2. 0. Bis 0.50090507 mg/mM 1.6 males SrO -> (1.6) (103.6194) = 0.16679104 mg/mM 1.0 MM GO > = 0.0795394 500- 5-CO3- 1.42472693 (0.16579104) = 0.2362 (0696) scale foctor for 50g lot 160 (60) (0.50090507)= 30.0543 Bi (0.23620696) = 14.1724 Sr as sico3 (0.0796394) = 4.7724 48.9991 Cao, 86 Sro.14 Cul Oz 0.86 (56.0794) = 0.048228284 (1.785.) = 0.0861 0.14 (103.6194) = 0.014506716 (1.42472693)= 0.02066811 1.0 (79.5394) = 0.0795394 scale factor for 50g butch (340) 340 (0.048228284)= 16.398 4.932 7.027 (0.02066876)= 50.468 g (less (U2)



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132.90

90.49

9/13 Run aborted. Restort. Col I @ 8000 then x-ray,

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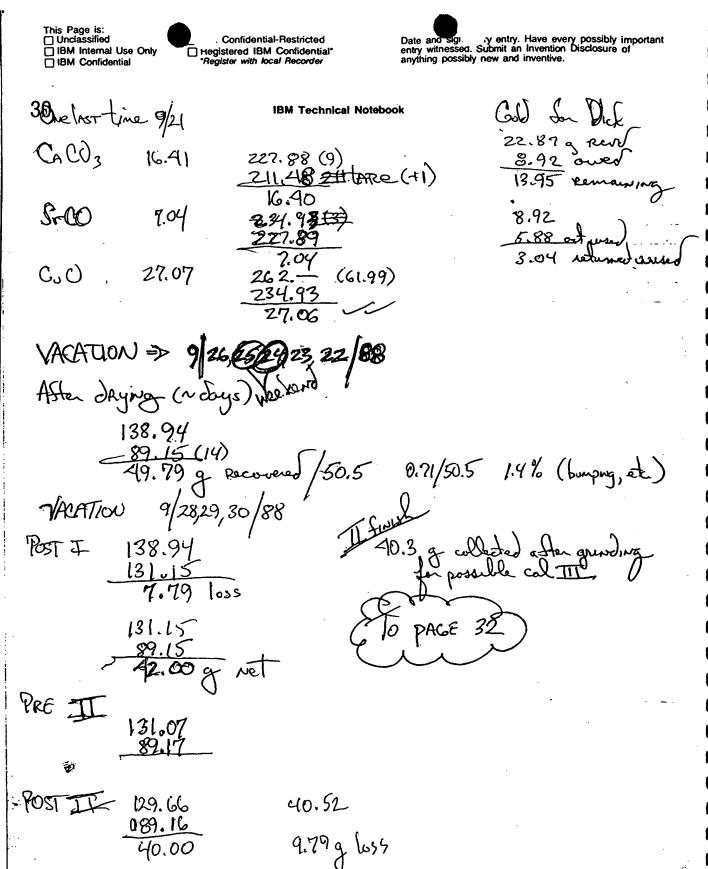
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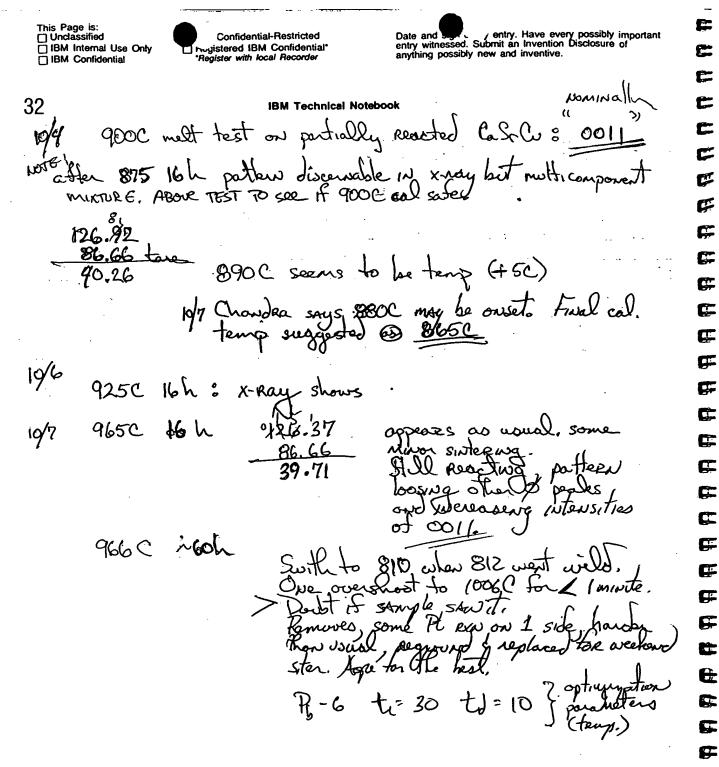
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34 P2 - N4600	150-28,500 IBM Technical No	tebook	
	•	2 4.465 <b>69.</b> %	
START SINTERIA	G @ 4:00 M Rope) -1	emp setting 835. Should give	e
	4:20 840 E 846 To 4:30 839 w	some > Ts 85\$ ? shalt	2) lange
	4:30 839 w	so Ts 859 Redsod	~ <del>~</del>
10/5	4:50 % 8:30 16h s	Stalol @ 856	
			2
Pellet warped	flowed (Sapprox da. du	surface. Reale temp as for exp below 850 for ext. (	Deg-
PRE as I saw.	was 859. Must ke	rep below 850 or at). C	- 0.
0.99	$\sim 0.21$	4,40 69.5	
	ood @ 852 efter?	20 h (overente) Keep sustan	7
10/10/0.90 1.23		3.3 51% does not make	
	tys people have seen	such effects usual H	wigh
Be 1, 68 ar. 17 Cu2 Op S "22.12"  Avalysis Results Pg. 33		10	noox
"22.12"		8	20h 250C
Analysis Results			
7			
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RE ~(1.1) \$ .13 1:098 68.2 J.257 0.27 65.9 0.261 0.271 W1.108

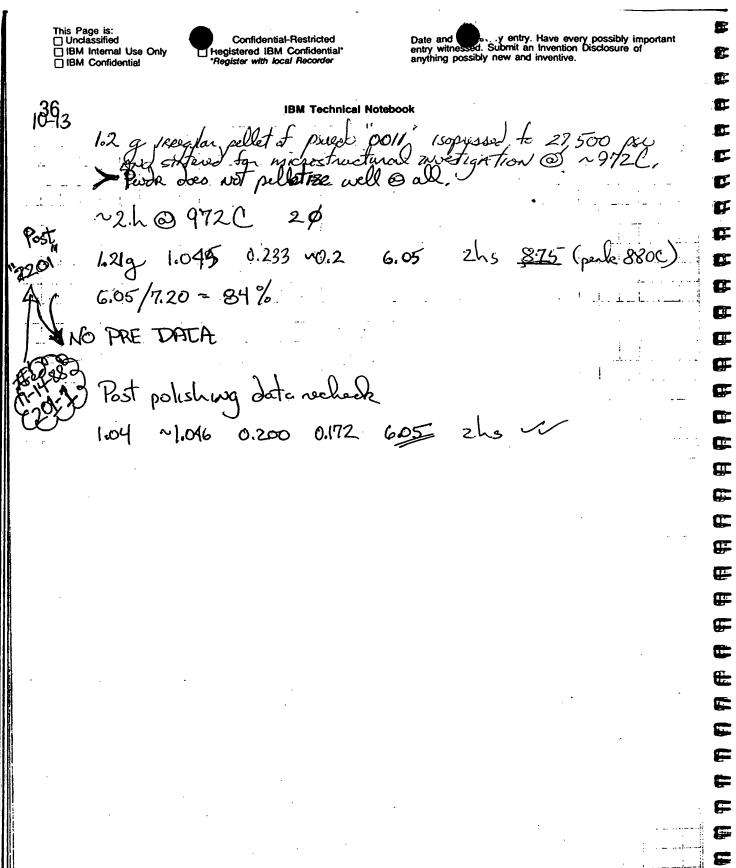
2(32-> W 2:34 >> START 6 853 2:34 > AUT

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Date

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37

**IBM Technical Notebook** 

Call3

Srcoz

29.27

27.04

Sille

after mixing { daying: 63.15/63.317 = ~0.3 % loss

29.29 (0.5603)= 16.41 7.05 (0.7019) = 4.95

Oz Caux: 48.1/48.39 looks complete

(47.84)/48.1 = 0.5% gading loss (to temp (966 @ 4:00 p.m. 10-19-88)

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38	IBM Technical No	otebook	
Note: 810	optimized process po	orameters	
			E
13-29 ti	= 15 td= 5 AP:	= 2.0	•
1	V 1	VARIAN	Xer I
B. Swithesis	B <sub>1</sub> +3% -	1 2212 +8.84%	· · · · · · · · · · · · · · · · · · ·
-C+0-169	Sc -6.25% -7.17	% +5.95%	
365	C45		
		A \ 1.0	· · · · · · · · · · · · · · · · · · ·
	@ estrated: 0.821/0.86 +: un	maked to Comment	
1	B. 2,22/2,15 -	- 2.34/2.15	······································
	Sc 15/16 08/	(H) 1.78/1.68	g
	1/- 08	%.%12/1.17	
			C
			9
	2 1		<b>9</b>
	HNALYTICAL Kesults	0011	<b>Q</b>
	B. 2:2K5) 2.15	act. thent.	
	Sc 1:5 1.6	0.13 0.14	g e
	<u> </u>	0.86	
	<u> </u>	1	•
	@ Nate: Concentration	of the to conspion of	
	code to contracte	Two so consultat with	, Nº .
	2212 large amount of	CO second po	
	<u>B</u> 234 2.15		
	2c 1.38 1'78		
	C		
	Cu Lo 2		
The above understood	Date	and	Date
I the man a minimum and		•	

10-26 DOM miled (19/25) & KROY Workerties & Single of 39

die body: 0.483" / 1.228 mm I.D.

(10) 8,500 psi purde press too Shooule to go in iso. left after a few attempts (10) pressure us knowleast orumbing.

Next time: ~ 3,600 psi => 16,000 may need to reinfly PSD with available presently.

Pres data with taken!

Pres data with taken!

(peole-5 min- (1866) Reput temp finds (1948)

1.36 (pellet damage > ~1.4)

3.68/4.86 = 75.6 >> 76. I damage)

1.174 0.352 0.381 cc 3.675 NEED pyrometer devery.

Sintered microstructure revends ~ 80-86% dever pellet gy muon 1-2% probably CO phase in some traple points. Grows 2 2 2000 monthly Coloners in some traple points. Grows 2 2 2000 monthly Coloners in some traple points. Grows 2 2 2000 monthly Coloners in some traple points.



1000X, 3h sinter, 0011, POLARIZED

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# IBM Technical Notebook

7`	VIII. DENSITY MORKSHEET		•	• •	VIII. B	EHSITT_MORKSHEET			-
		STEREOPYCHOMPTER	4.95 th	enetical			STEREOPYCHOMETER TRUE POWDER DENSITY	-	••
	EMPLE WEIGHT 4.06	q. OUTGASSING COM	D DEFENDES ALL		SOURCE TOTAL TARE W	MSICHT 14.04	- OPERATOR	-27-88 PRD DITTONS N <sub>2</sub> V <sub>2</sub> 85.72 ec  LONE, V <sub>3</sub> 14.65 ec	
	OPERATIONAL EQUATION V <sub>p</sub> =	$V_c \cdot \begin{bmatrix} V_A \\ 1 - P_2/P_3 \end{bmatrix}$			OPERAT	TORRE EDGELLOR A <sup>b</sup> ·	$= v_c + \begin{bmatrix} v_A \\ 1 - v_2/v_3 \end{bmatrix}$		
	$V_p$ = Volume of Powder (cc) $V_c$ = Volume of Sample Cell $V_A$ = Added Valuese $V_A$ = Pressure Sending after	Holder (cc) or Presentising Coli	•		v <sub>o</sub> - v v <sub>A</sub> - s s <sub>A</sub> - s	blum of forder (cc blum of Sample Cal dded Volum resoure Bending oft resoure Bending oft	l Holder (cc)		
	3.685	369(S) 267(S)	3.491	3692		3.568	3.5 <u>1694</u>	3.54	<b>196</b>
	PON 1	MOH 2	<u> </u>	Roug .		pare 1	ers 2	<u>808 3</u>	
	19.646	19.831	19.683	19.718	P <sub>2</sub>	19.865	19.720	19.807	12,661
	· <u>5331</u>	5.372		5.841	P3	<u> </u>	<u>5.530</u> J.522	_5.83	5514
	· 2.999 ···	3,0% ~	3.07 €		₩,	_1.548 <u>~</u>	- 3-35E "	1.522 00	
	DEDISTITY 494 4/00	4.86 0/00	4.87 9/00		OCHSE	**/cc	7.21 0/00	7.21 ./00	

The Tourse (aguel. values) rete: prefected chamber and overshoot 1886 950 978

1888 948 977

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Date

#### **IBM Technical Notebook**

Pission Pellet Calculations:

123 d) pellet volume: 3.25g/6.36g/cc = 0.51 cc

2201 = 0.51 cc x 7.2 g cc = 3.67 g ~ 3.75 g

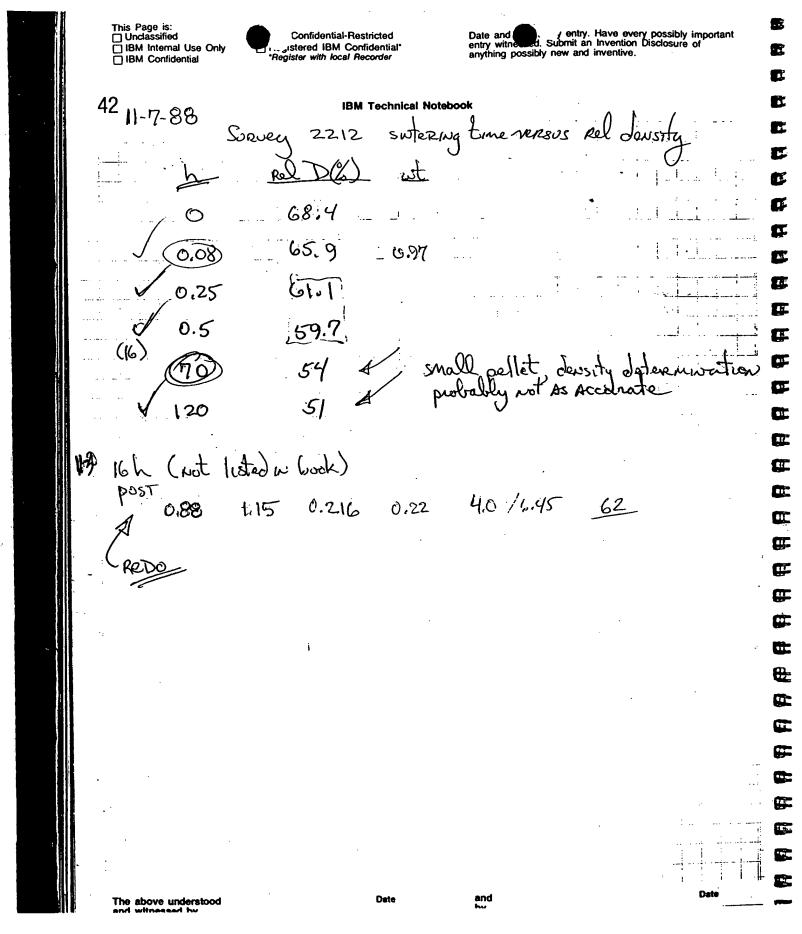
0011 = 0.51 cc x 4.86g/cc = 2.48

Two preheated RT @ 3:31
The set To Tomple
9148 92? 933 3:31
956 965 3:33
958 964 974 3:35

950 956 974 4:20 951 4:21

backing off now to maintain temp. tell EQ

41



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	10-2-88 0011 E1. Wt %_ Ca 22.4 Sr 8.24	Analytica BM Te the Mal Aval 0.86 0.8	875	0.639/.634 =	oney From
	() 40.6 11/3 Pre 4000/30,000		Srud Caudi Caudi	2 : ( = 0.539 = =	; <b>-</b>
	2.85 1.531 C	0.496 0.913 0.639 0.64	~3.12. /	4.95 = 63—	perdect
	2201 4,000   3.78   1365 (	9.494 0.723	5.23		(too high?
	& Post 875C for 301	colluse for first	press varsed, grown	Jarge volule	
	provious 3h s	nter stoward w	o europeuse of rus	lability.	
M.A. A. A.	p.			· .	
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2013 2.99 1.35	0.415 0.595	5.02	69.7	alen bansipi
2.96. ~1.30	6 0382 0.498	~5.9 <b>8</b>	69.7 ~ 83.3+	pychometer 86
			PATTS MW	ar= 85
2014 1.16 1.082	0.245 0.225	5,16	71.7	6 min.
1.15	2 0,245 0.225 5 2.10 0.182	6-32)	87.5	double of
		:	7	
2015 1.17 1:080		5.09	70.7	Stroken bestore
 	reground & P	Det 201-112		
-	O Representation			
29/16 0.99 1:09/	·	5-13	71.25	to temp 1845
*† 0.98 1.06	0.185 0.163 N(0.178) 0.158	6.01 6.2	83.5)	36 mw)
		رامه .	De	-
X(f): 201-4,6 so	me eurobrice of drooping	og A	in pellet.	reduce temp 5C
2700/29	0.25% 0.238(5		69.9	to temp 2:27-8
101 2/06	0.185 0.36			2143
1.06 1.19 1.05	~ 0.225 0.195	6.1	84.7	75
				# · · · · · · · · · · · · · · · · · · ·
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3 3 6	11-10 Sistering	Summey 2	ical Notebook 20	201-2
	201-8 87 <b>24</b> 201-3 872 201-4 875°C	2 min 71.25 12-3 49.7 <sup>†</sup> 5 71.7	85.64 OII 86 4 87.5	-2201 P3 pressed pullet large die
<b></b>	201-9 <b>87</b> 2C	5 71-	84.7	
3	201-7 <b>8</b> 72C	15 69.9	84.7	
<b>33</b>	201-6 875€	30 71.25	83.5	
and a second	201-10 8720	14 70.4	86 ?> 85	
	201-1 875	24	84	
	201-2 875	30/	<del>(16.1)</del> 79	
1 0 1 1 0 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	• •	معنی کارند ا		
	Record beaping:  PORT  BAS prevonetry  (wght 4.6g)	201-2 30h 875 201-3 0.608 dia 201-5 regid => 20 201-5 regid => 20 201-5 regid => 20 201-5 regid => 20 201-5 regid => 20	of ~ 75% (pyc):  thing in ranging he  political for press  1-11  A Rol density for  84.75 (Reosonable	replets 1,78,9  agreement), mostly
The state of the s	The above understood and witnessed by	Date	and by	Date

19-14	SINTER	めつい- タスくつ	Technical Note	73:4	y *		1· D
70148 W	ect pellet ~	3000/290	oo Linu	inter timi u: Eq soa	ES ARE 1. K+DSW	nw attain TER time	mont 7
1.19	1.081		0.232	• •		green	
n 1.17	1.038	0.225	0.190	616	85.6		
201-9 12	: 1.075	0.259	10.235	Sill	71 <del>00</del> n	green	
Lo19	1.036	0.231	0.195	610	84.7	1. 1.	
201-101.1	0 1.083	0.236	0.217	5.07	70.4	أجعدا	Code
1.09	1,413-1.07 M <sub>1.0</sub> 57	~0.2+	0.175(	5) 6.21	A SECRET	(accura	te?)
201-11	Pallet	bsper coy	& repressed	Snonfa	oku pellet	Also, die	RAM
· ·	1.169	0.252	0.27	5.04	olen pellet ressure (d. 7 70,00	we kag.)	2,000

? probably slightly less due to exclusion of sidge volume and liverage after flattening; 15 um

201-10
0.90 1.057 0.168 0.147 6.12 85 better (more of 100)
10 2.96(18)
201-11 1.357 0.412 0.596 4. 496 69.4)

10 2.935 1.3150.365 0.496 5.94 82.5

The above understood

and

11-12

IBM Technical Notebook

47

201-11 cut what larger flattened and polished.

0011-2201 sandwhich ~0.353-0.363 thick

> From furnace top to bottom of "weight plate" 1932 @ 462C assuming ~ 6 lbs for ram & plate & x-sectional pellet area of 0,212/N2 load 28 psi

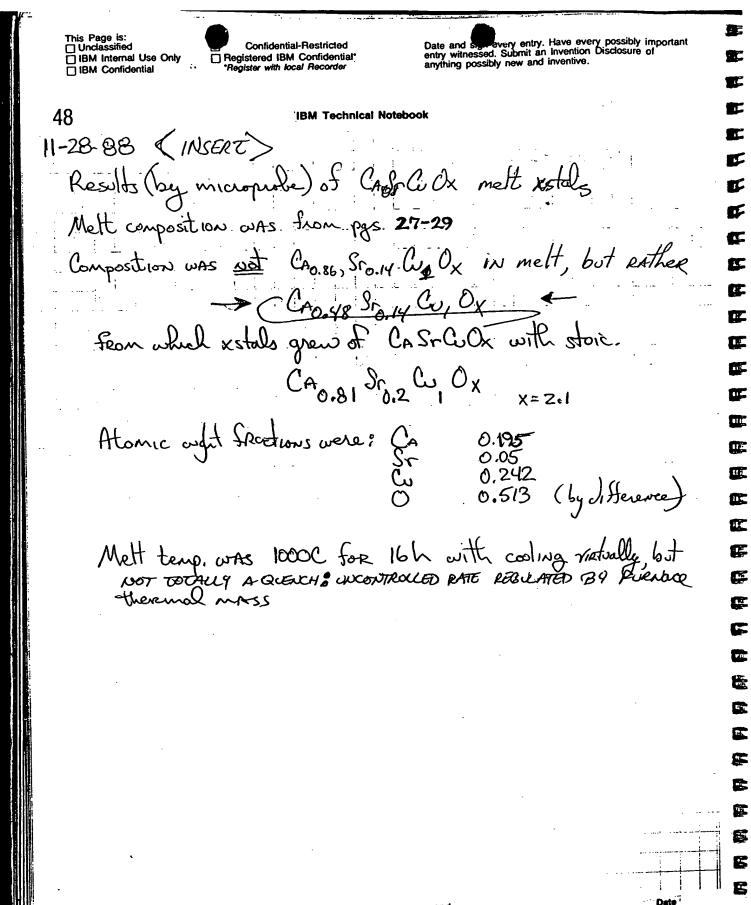
Tdissussion sistering set @ 8600 for ~ 12 hs.

Rel dansity from measurement of 201-11 ~83 %. On suspection, of interval poliched sustance numerous buenout-like occlusions present. Some dagree of open porosity, also.

Pyc. pol. den = 38. % thus 1 attachetel to open porosity.

0011 pel done from messure ~ 89%. No pyc reading done
16 h sinter @ 9750.

41:30 pa. TO 859C assume start of diffusion sinterink Plate hight 13/8" (3/32 exponsion due to TCE from 462CD) No Rt measure mode, but not significant



11-22	IBM Techn	ical Notebook		49
Balance Bi	Pudes Son ?	Rxw		
2212 - 30.5			7 · · · · · · · · · · · · · · · · · · ·	
2201 - 12.5	· · · · · · · · · · · · · · · · · · ·		:	
0011 - 33.5		1 1		
2nd Diffusion	Run 2he RA	mp to 866C	@ 1000 pld	e spoce = 1 7/3
2201 n Same E	) @ 018 cm (	at measured whe	while and	they was
2201 ~ Same C	). <b>\</b> \$		,	0 11
loose ~ 0.23- so sandonud	0.2 × 0.03/s/ke might be ~ (	0.63→0 0.29 cm (80)	( of Run#1)	· 0.19
a grow thuleness!	my hought of plate	gitterences @ 8	66( 1 1/32-	1 732
\$32:0,23 cm.	· . 189-0.23 = 0.	06 too small	≥871c.t	zak
RESULTS: "Bi the Lia per	pellet has spran	appuently m	etting. lot	al
La	phery.	talline (3) skirt	t around pel	let n svetere
•		O =	hottom Ram	
		top sole	cos .	

0.18cm = 0.07" slice ~ square 0.07+ 0.015 = 0.085

The above understood and witnessed by

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L	ſ	1
7	1	1
v		J

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## IBM Technical Notebook

50		IBM T	echnical Notebook		
0011- 0011- 0011			By Srz	CA CU, O6 CA, CU, O2	
X	+ 5	<b>5</b>	ر مرا <sub>ره</sub>		
From	ideal"	stac.	2201 + 001	1 2212	
BC	1.W. 208.98	0011	22.01 417.96	2212 47.96	
S <sub>r</sub>	<i>8</i> 7. 62	• • • • • • • • • • • • • • • • • • •	175.24	125.24	
Ca	40.08	40.08	_	40.08	
$\mathbb{C}$	63.54	63.54	<b>63.54</b>	127.08	
0.	15.9994		95,9964	127.9952	
•		135.6188	+ 752,734 =	888.3562	
BC		0(014)(086)1	(2.15)(1.6)(0)(1) 449.307	kisX1.69X1.19)(2) 449,307	
26		12.26.8	140.192	147.2016	
CA		34.483	-	4/0.3284%	
$\mathbb{C}$		<b>63.5</b> 4	63.54	127.08	<b>,</b> 0)
$\bigcirc$		31.9988	~95.9964 <del>*</del>	X127.9962	9997 (o
		142,2794	749.0354	891.912296/	(891.398)
% Sv	prieratore	+8.6%	*(5325) 519994 93,1965	99.6% (2	2 ilselz
I ne above u			bor bor		

51

### **IBM Technical Notebook**

CONTINUATION . . .

1 mole "0011" + 1 mole 2201 = 011 + 529 at % 2201

142.27449 + (0.02) (342.2744) = 145.12 g } 2 at %

142.27449 + (0.05)(142.2744) = 149.3881297.11372 = 9.96 $9.485 <math>\sim 0.475 = 9.96$ 

For Stoic (mulan) Mix = 1.423 g + 7.527 = 8.95 g botch size

Total Verge 0011 2201 %

20.393 **8.192 6** 

vol 2.37 0.0264 1.0% 2.

table 2.37 0.0736 3 5

STOIC: 385 Sr Ca C

STOIC: 285 Sr Ca Cu
2201" 2.15 1.6 0 1

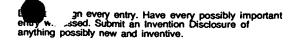
2.15 1.74 0.86 2 versus poly 2,15 1.68 1.12 2

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and

Date



52 Stoic MixING

**IBM Technical Notebook** 

0011 2201

~1.43 ~ ~ 7.53

MIX STARTING @ 3:00 P.M., 50 mls 150 Reyl. 5 cc 2002 balls 2/3 full

NOTE: From pottom pg 51 can be seen this Additive approach will yield a theoretical mular comp & O.1 M larger in Sr O.31 M less in CA

1.e. Sente en Calcia poop

8.96 g assed witially, 8.85 g recovered's 1.2% loss (98.8 yield) Stor 1 Fre 2700/27,500

3.11 1.36 0.486 0.706 4.41 ~ 689 0.25 (4) + 0.75 (7.2) = 6.4 vol % basis, ~ density cole

Rw. (SINTER) temp to be 850C

Relet metted indicating lower mp lip & exists in system of later testalling. Prodomenantly 1 lath-like & my exigen when growth as in 201 120h sample.

53

12-5

4:20 P.M. 1/4:25 @ temp\_

No per data on dessity due to irregular shape consedling pellet crimbline during isopressing.

No per data on dessity due to irregular shape consedling pellet crimbline during isopressing.

Unipress > 6000 ) 150-29,000 PSI with ~ 3.19

12/6 9:35 Slow cooling begin . DTenter = 17h @ 8750

Post 2.869 ~0.460 mm thick postus myst have been ~1.380 estimated daysity 0.666cc @ 3.19 ~ 4.65/5.00= 93 (may be high) 3.0 4.5/4.5/90 better

Slice 1 + 0.09" often dearing / post polish > N/R

2201-8 1.088 Sia is asea = TTD/4 = 0.85 cc² = 0.409 = 0.525 m² 5.75 lbs/.525 m² ~ 11 psi

2201-8 (top)
Pellt consignation @ START ~ 3:55 p.m. thickness - 0.34 cm
201-3 RAMP > 434 Set point - 800C Duell-12h 132@380C

12/7 Result: no meltinge, pellets bonded of little detarmation.

12/8 After 24h 3250 Anneal no evidence of lie., but bond breaks afterhanding at pellet interface with some "Axiv etching" of coll pellet surface leaving thin, layer of 2201 (or exist prod) behind.

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Date

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This Page is: Unclassified IBM Internal Use Only	3M Confidential-Restricted	Date very ententry witness J. Submit anything possibly new a	try. Have every possibly important an Invention Disclosure of nd inventive.
52-6 SECOND 22	IBM Technical No.	BL 2.15 Sr 1.6 CA	0 1 W, Oz
i	30.0543 × 2 14.1724		
:	4.7724 48.9991 g		•
	ersion Lator For CDz	·	
Estimate ~ 89 g	batch recovery	97.9982 - <u>8.4468</u> 89.55	_ Cd <sub>2</sub> /033
12-7  Ear 202.54  By 03 262.68  60.13	60.11= 10.02 /		-
SrW3 291.03 - bore 262.68 28.35	- 271. 28.35 wyb 28.3 D V	6 tunhad 28.36	• · · · · · · · · · · · · · · · · · · ·
201.57 291.03 9.54	(300.57)/9.54) wyb 9. D	55	
12-8 97.9 <b>2</b> /800	overy after Leying ove 22 theretical =	earle 99.9 % yell 0	.1% mixing 645
to	£36-7	. •	
The above understood and witnessed by	Date	and h	egi, volgend i <b>Date</b>

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**IBM Technical Notebook** 

55

9.49 0.48 theor.
9.48 0.50 which
0.49 color o.49 actual = 0

Std. 1.-1.5h 5min ZrozfIso grind MIK, scheening & drying.

12-8-85

Recovery: 9.84g/ 9.86g theoretical = 99.8% >012% 1045

COST TARO 51.04/5 9.83 teassered

0011-2201-5W(3V)-1 Post 8500/29,000 2.31 (117 0.704 0.690 3.35 ~67%

Pellet dadeger than usual, 1.75 g max in Littre might be considered.

12-9 5W-Z 900C 8500/39800

1.27 1.174 0.382 0414 3.07 61.4

3:55 N perheated fuerace + 4:00 to temp@ 900C POST 5 MIN 1.111 0.36 0.349 3.55 71-

15 MW NO SIGNIFICANT CHANGE

12-12 to temp ~ 10:30 A.M. (chee: 10:45 > 40 stumping) > SWIER till 12:130

1.24 1.055 0.33 0.29 4.28 N86%

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2201 SyNII cont. (fun py54)

cruchle tore \_88.79 97.99

10:00 AM >> 575C hold 1h

11:00 AM

cool, regulared to <100 ment

88.95 (well other sistened public body removed)

93.99 if 88.79 vsod
181.02 after gounding
88.95
92.07 to temp. (866 C) @ 1:00 p.m.
- 1.21 in genting
1.3%
97.88 - 93.28 = 4.61 } 55% RECCTED

1:00 - 5:00 pin 866C, shut down for weekend (may restort son ever)

12-10-88

to temp 866 @ 10:00 A.M. 12/13/88

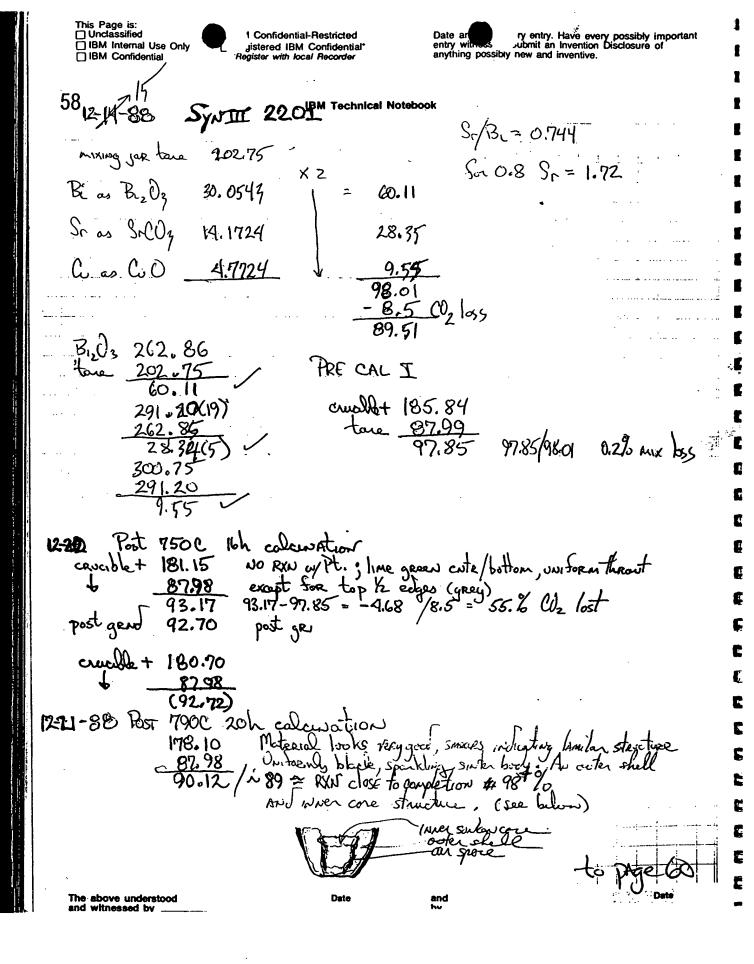
PARTURE MELTING, "classic" Extertic lamillare And longe 2201 lather.

The above understood and witnessed by ......

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Date

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12-14-8			IBM	Technical No	tebook		59
	Y1 BA2	Cu3 0x	Impla	tation	Experia	IENT	
R	E- Silm	on Stills	3500	30,000	160.4 %	, ·	
3.07	<i>\$448</i>	0.485	0.799	3.84	160.4 %		· · · · · · · ·
5	<b>!</b> !		**	•			
: 		a line mas	KER 1	o long Axis	s of trimique	by Solida. 3	inplant
		- implime			MARK ON ON	DERSIDE OF	pellel
3.02	1.271	0.3%	0.508	6.02	94-95	opposite	
ti. en en		··· · · · · · · · · · · · · · · · · ·	· · · ·				
· · · ·	·						
34 · 4 · 24 · . · · · · · · · · · · · · · · · · · ·			· ·	***			
3.05	1.448	0.476	).784 3	2.89 ~ 0			
+	(OET)	~ lae m	exer 11 to	is long cut	two Axis of	Two pellets	(cut on live had such
		- tralane	70,0,L. L.	1 - 5005			$\begin{pmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & 1 & 1 \end{pmatrix}$
2.99	1.272	0.39)	1497	6.02	94-94	E POLO	had slee orang
3	•		·				
	•						
5.10	(15)		.a.1 1	V4/	10.01.1	<b>—</b> .	_
	_	5C @ 10	•		, ,		N 6:00 pm
Citting	125193	: trodam.	measures	0.5° on	SAW (O.	085-0.505	-tougents)
1.222	54 - 0. <b>4</b>	Set J				•	
				•			
	ve understood		De	ate	and		Date
and witr	nessed by				by		Deta

by

12-27-88

**IBM Technical Notebook** 

61

Sommary various RXN pellets:

5 wt % 2201 in 0011 for 16h @ 850C 5EM

5 wt % 2201 in 0011 for 2h @ 975C 5EM

[0011-2201 STATEM MINITED pellet: 13h 850C low prosecuted graw growth/warpage] both

100 & formation, exagginated graw growth/warpage

0011 @ 975C 17h sem 5TD.

2201 @ 875C 1h sem 5TD.

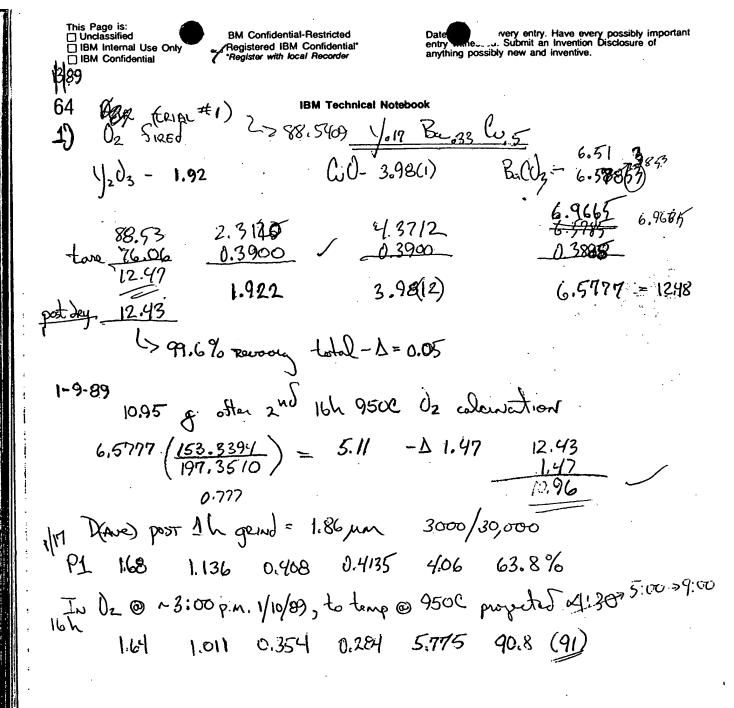
2212 @ 853C 5 Min sem 5TD.

•						
62		IBM Tech	nical Notebook			
12-29-80	DAVES	Composition	ΝĞ			
# )	Bu	C	()	$B_{\sim}$	Cu	·
(0.1 <del>67</del> )	(0.33)	(0.50)	0.17	0.33	0.50	
0.15	0,33	0.52	0.8634	19038	3—	
0.17	0.35	0.48	1.0625	2.1875	3—	·: - ·
0.19	0.33	0.48	1.1875	2.6625	3—	·
0.19	0.31	0.50	1.14	1.86	3-	
$C_{0}$	to Como	sitions (cal	lanktions	Next Down	2	
	190 Congress	B~*	C	total		
1	1.91937	6.51253 (6.51)	3.97697 (3.98)	12.48	*	
2)	1.69356 (1.69)	6.51253 (6.51)	4.13605 (4.14)	12.34	A	_
3)	1.92	6.90723 (6.91)	3.81789 (3.82)	12.45		
$\Rightarrow$	2.14518	6.51	3.82 0.48	12.48		,
分	2.15	6.1783 (6.18)		12.31	K	
* B	as Bac as Y20	03 } NOTE:	De purity	Corrections	appledy	et e

IBM Technical Notebook

63

kulations for whits summarised on page 62 2) 40.15 Bao.33 600.52 Ba= 0.33(197.3434) = 65.1253 q (29.5394) = 41.3605 q CV V= 0.18 (225.8082)/2 = 19.1937 g y. 03 ) 69.0723g g BaCoz ) = 38.1789 9 60 4 0.19 Ba 0.33 Couls Ba= 65.1253 G= 38.1789 Y 0.19 Ba 0.31 W 0.50 Ba = 61.1783 C= 39.7697 65.1253



IBM Technical Notebook

Sa 0.33 Woots

~ 2.15

5

6.9613

4.2017 0.3865 3.8218 3.8217 ·

نر.2082 علام

10/88

January STARTED affar cending. No officeration. Purch looks good allowardy.

10.99 a offer 2 nd colonation;

10.76 post grind

We 6.58 (.777) = 5.11 -∆ 1.47

11.00 g expected: 1099 1 11.00 g expected: 1099 1 to temp @ ~ 5:00 p.M.

VM P1 PRQ

2500/30,000 0.399 0.4073

3,93

62 /0

1.5%

1.60

1.05 v 0.365

0.319

77.8 %

Green of peaks coming up in X-RAY.

The above understood

Date

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**IBM Technical Notebook** 

67

2201 P1B3 1.09 0.391 0:365 1.94 15mm 860 0.359 0.313 494

2 ut % 7201 N 0011

9.49

some bealing during 8.5 h mix : 9.05 g

4200/30,000 to temp @ ~ 5:00 p.m. ~ 64.2 (80.25) 2.82

.69 ~1.21 4 1.194-1.227 some slumping 0.525 0.60

-A 12%

1.57 mm 1.12 mm 1.79 mm 0.25 0.380 (+w)

F F

E

C

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E

68

## **IBM Technical Notebook**

2)  $\frac{y_{0.15}}{y_{2}y_{3}} = \frac{B_{A0.33}}{3} = \frac{C_{0.52}}{B_{A0.33}} = \frac{C_{0.52}}{C_{0.52}} = \frac{C_{0.52}}{C_{0.52}} = \frac{C_{0.52}}{C_{0.5783}} = \frac{C_{0.5783}}{C_{0.5783}} = \frac{C_{0.5783}}{C_{0.578$ 

bittle tous: 74.55

(.1270'5) 1.920G (.1270'5) 1.920G

6.8066

17 ST CALCINATION

tore 54.16 //12.41 = - D.3%

6.5783 (.277) = 6.111 (-1.47) 12.37

~ 10.90 expeder yeld (less tronster losses)

1/18 post 66.53 -5.15 (05) take 54.20 Recovery 65.05 -1 1.38 (48) TO.85 Ntotal RXV.

1/19 POST 65.02

54.2 10.82 / ~ constant 10.29 secony

Notes: large lia stains (formation) during 151/2 nd cal white

30,000 750 5:16. ten @ 7145, 16h & 11:45 AM

3.95 62.1% 1.474 0.258 0,405 1.60

1.227 0.216 0.255 6.16 96.9 1.57

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\*Register with local Recorder

1) 0/2

4)02

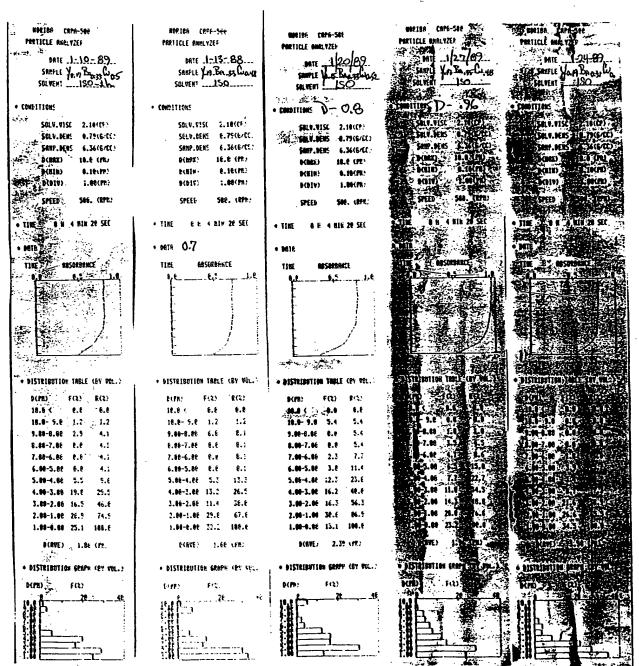
1BM Technical Notebook

2) 02

3

\$ 1 h iso 3)02 PSD 5

69



## **IBM Technical Notebook**

Sry W120 -> Sr0.37 C 0.63 ON	5,0 38.34	C.O 50.1078	
S.C.O -> Sr.0,5 Lo.5.Oz	51.8097	39.7697	
Sc. (10) -> Src. 67 (1033 03	69.4250	26.248	
SrD = 103.6194 -> SrCDz H	7.63	14247	
3.83 6.02 3.834 5.0198 -> 8.8538 (5.18) 5.1807 3.97297 (78)-> 9.1579			
(a) 6.9415 26.248 (b) -> 9.5673	······································		
5.46 5.02 10.48 - 7.38 3.98 11.36 9.89 26.34 12.52			
S. C.			

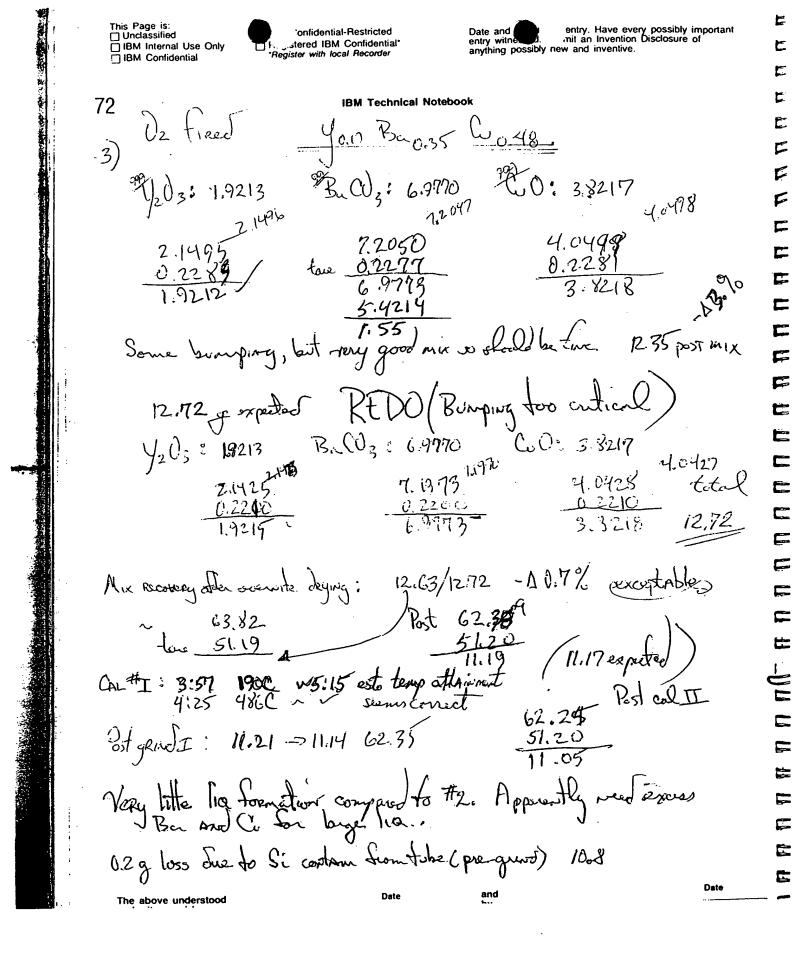
| Shice 2 - 1.28 mm - 1280 mm | Shice 3 - 0.68 680 mm | Shice 2 - 1.28 mm - 1280 mm | Shice 3 - 0.68 680 mm | Shice 2 purps; nowaked side 1 measures ~ 27.64 | 26.30-(3)).

aum: 300 m | 1340 | 26.30-(3)).

aum: 300 m | 1340 | 134-1.29 |

720 m before starting second side | 150 8's on 6 give 770 |

720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 150 8's on 6 give 770 | 720 m before starting second side | 720 m before starting second side | 720 m before starting second side | 720 m before startin



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10.97 collected: white top layer on powder. Day Slake-cake very agalamented / buttle and does not easily puch out when Had to day gund morder to produce deept

3500/29000

0.266

0.424

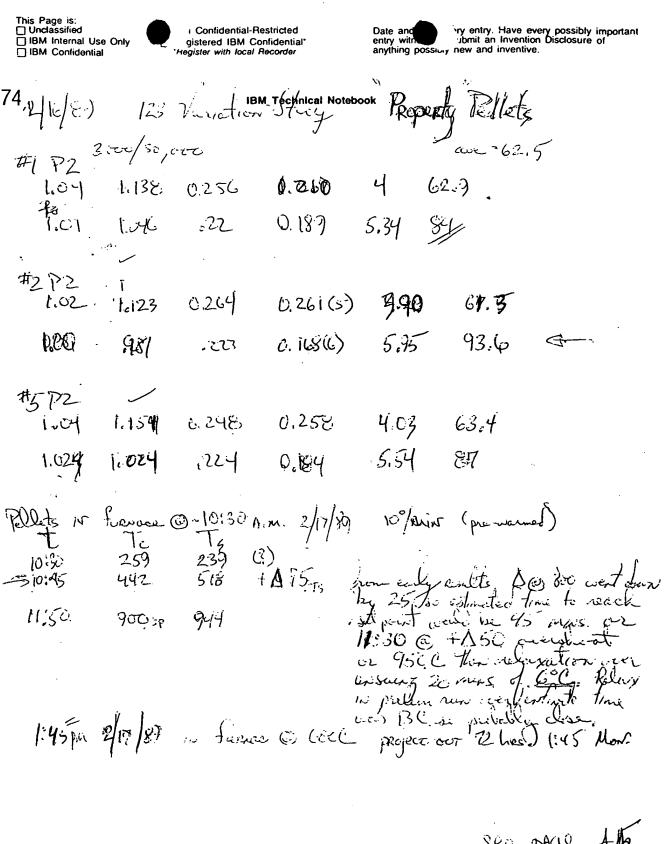
1.52

0.245

Pregund { Post grand x-rays show charge of some peaks IN two x-RAGS, however sintering colonation of pollet may return the products to original of; Will do x-ray of pellet

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Date



see page All

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268

**IBM Technical Notebook** 

75

Clean gas with Assenti.

Clean gas with Assenti.

Sample I deuse closed

poroxitoz, e 791%

cut sections from centur

Schryle 2 open poroxitoz

Center Sections

thin plies oxygenate

(1) I cut for ELSS (P. Batson)

(2) Magnitometer (T. Magning)

(3) Induction (Diane dinos)

(4) Cor evolution on disedition

1.85 intersect

1.85 intersect

2.3 925C 11

2.3 925C 11

2.3 925C 11

2.4 11

3.5 11

3.5 11

3.6 12

3.7 12

3.8 12

3.9 5

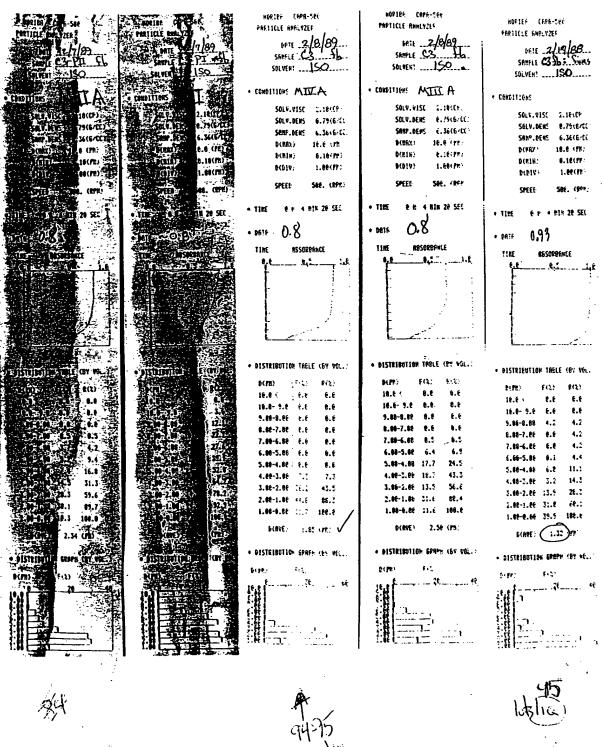
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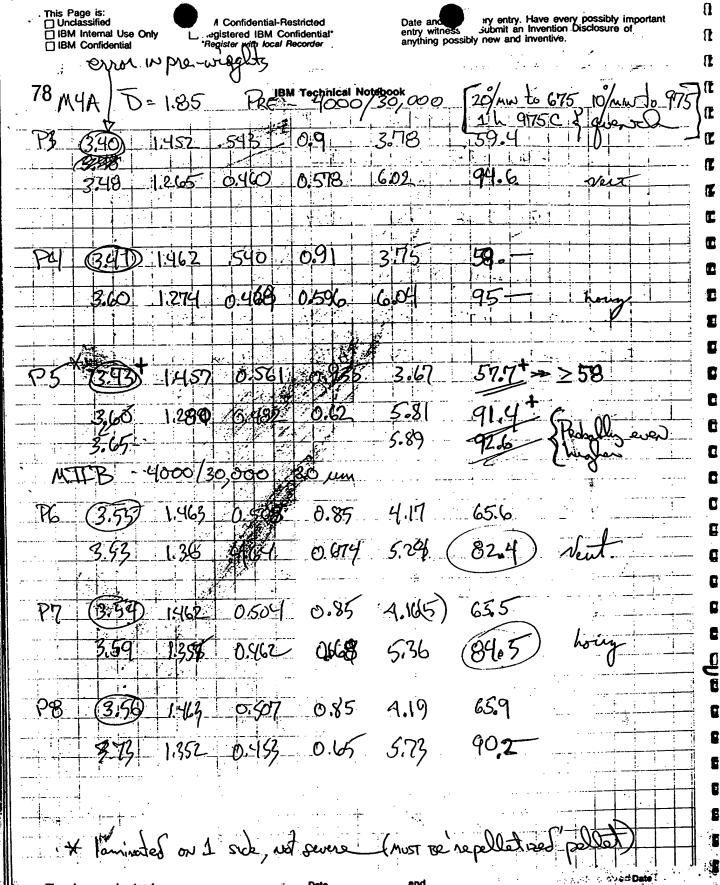
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<b>3</b> (	9113	IBM Techn	icel Notebook	•	79
3 Y 3	9 4000/30 000 3.57 1.462				· · · · · · · · · · · · · · · · · · ·
	3.57 1.462	0.517 0.81/	4.1_ 6		
3	3.56 1.359	0.463 0.666	5 1 5 5 5	1.	
	P10 Fines 4/3 0				
<b></b>	1.53 1.436	0,254 0,411	3:72 58	3.5 var expec	ted
	1.54 1.254	0.200	6.02 90	1.7 Spesie	lack pood
	Pellat culting NEXT	( see og A for	plan overview	)	
	Pellet 3 4 Jes	icated to ver	ical & horre	slicing	
	Pellets 647	7 7 1			
	from tangent:	saw cut edge:	1.2	ne 0.055 eg	<b>&amp;</b>
	Low Deugsty Vest	calulus i 1	1.2 show }	7 altogether + sx	Dece
	Hor	youtal : 2	1.0 spices mid-	setion	
			1.0 bottom		
	High lansity	VERTICUL: 5	slices (leve) lost	to chipping) len	I pholed
	~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~	na 2 3	oxy genater		
		hor12 : 2	mid section	(tox chipped)	· .
		·	.,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,		
		S. C.		and the second second	,
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TOP YIEW SOAK	DEM Technical No SIAGRAM  Pacements 4.142)  pacements 4.142)  pacements 4.142)  pacements 4.142)	Denne Discording Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents Documents	Min (10/h)
• •	Pollet (1) 0 & C.	19/mm to 850C, 10/h to	
,	2) I slice for Japan Brows 2) I slice for Alex for 3) save Ramander for 1 2A) I Atea slive oxygent Pellet (2) Of C ima horrzowial slices 44	organise (dessicuted)	
	1) I slice (10 mm) gound to the source in isopeopyl I disc to Tom 2 discs for TEM 2 spare	Ac (it possible).	
	Pellet (3) D&C  spane for (1) x-ray (2) (0) 2 0	lettree voldien	5
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	oxide atomic o coxide		
	1/. () : 0.17(5) = .17 225.81		
	- 12 U3 VIII VIII VIII VIII VIII VIII VIII V		
	B.O: 0.4(59) 33 153.34		
1			
		······································	
*	CO: 0.3665)~ .5 79.54	1	
	Go with the news	4	
	So write the local	pxid	e
	Transle Cale: at free doew.	56= 0.175	ROC HAN
=3	Valy 225.81 2 120 = 58.39 = 19.19 19.19/109.		
	BaO 153.34 x .33 = 50.60 50.6/	= 0.4618	3(5)
		, = 1.363	
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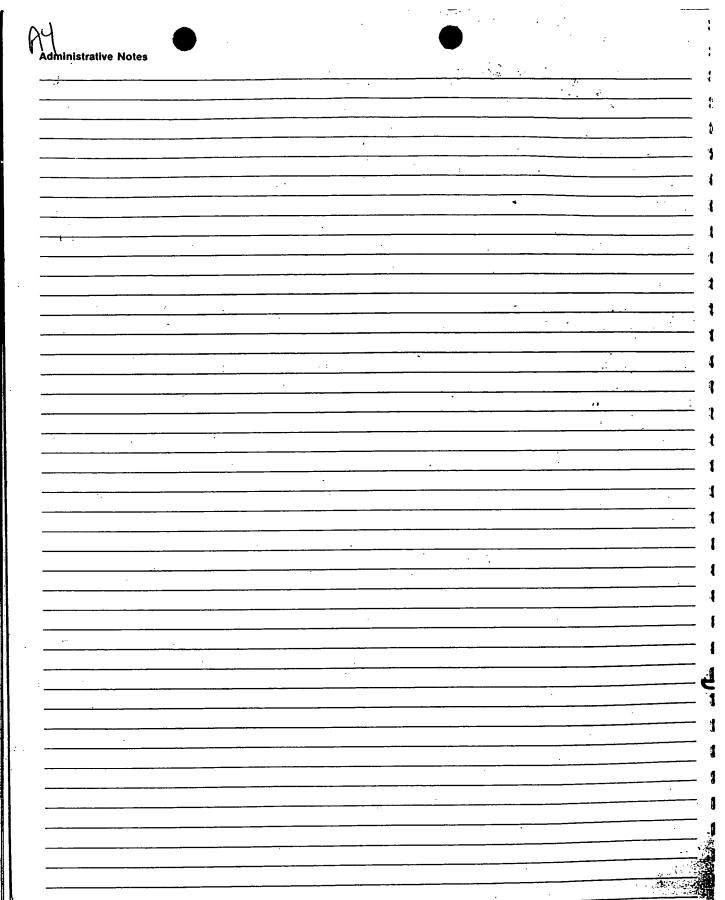
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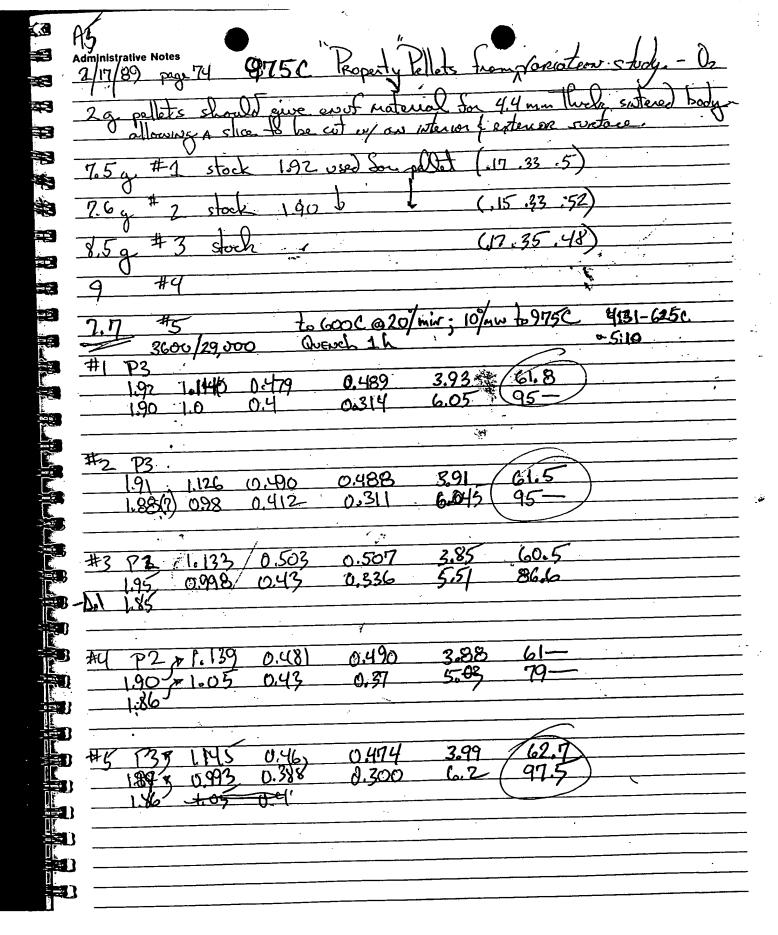
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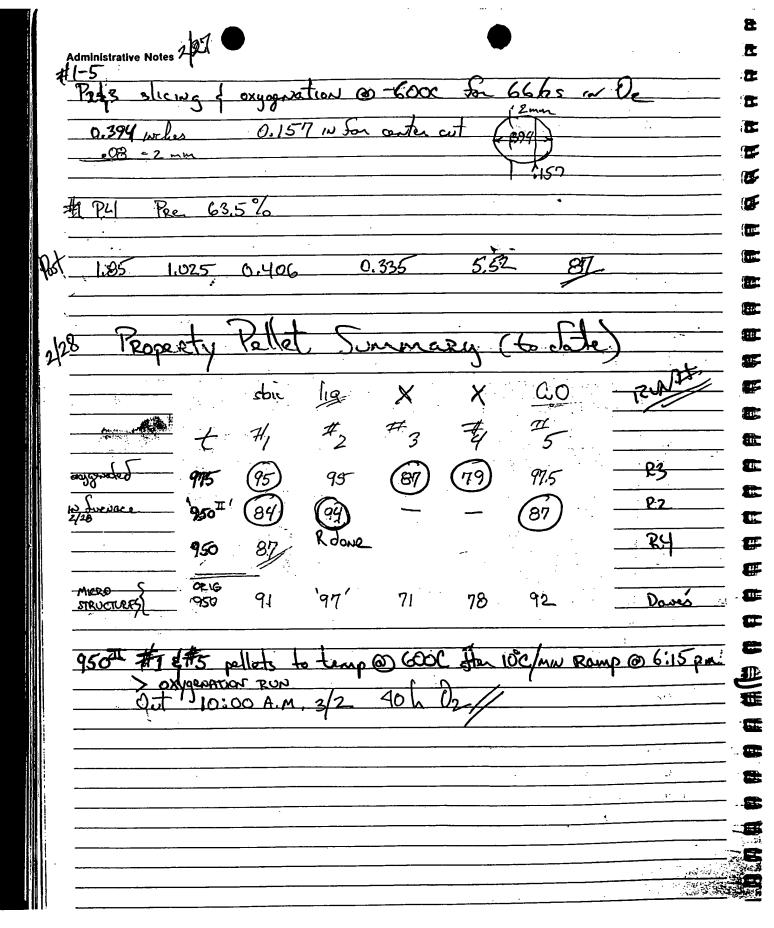
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. 4	77.8	4.95 62 3.93	
5	87 (91)	5.54 63.9 4.03	

\* after 1h swher @ 950C

Attachment G



### COPPER OXIDE SUPERCONDUCTORS

Charles P. Poole, Jr. Timir Datta
Horacio A. Farach

with help from

M. M. Rigney C. R. Sanders

Department of Physics and Astronomy University of South Carolina Columbia, South Carolina



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"A Wiley-Interscience publication." Bibliography: p. Includes index.

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tional spectroscopic evidence for lattice instabilities is not conclusive (cf. Section IX-E-1).

#### 3. Ferroelectricity

The high-temperature superconductor compounds are related to perovskite ferroelectric types (Cross), and the discoverers of the first such materials were aided in their quest by their experience with ferroelectric phase transitions in oxides (Mull1). The associated large dielectric constant may provide a channel for the enhanced electron pairing interaction (Chuz1, Zhon1).

#### F. SUBSTITUTIONS

Two of the high-temperature superconductor types LaSrCuO and YBaCuO each contain a group IIIB ion (Y, La), an alkaline earth (Ca, Sr, Ba), copper, and oxygen. An important question that arises concerns which of the constituent atoms are essential and which can be replaced by related or perhaps not so related atoms. We will examine atomic substitutions involving both  $(R_{1-x}M_x)_2CuO_{4-\delta}$  with copper constituting  $\frac{1}{3}$  of the cations and  $RM_2Cu_3O_{7-\delta}$  with copper accounting for  $\frac{1}{2}$  of the total cation content. Partial atomic substitutions and total replacements at all atom positions will be discussed. A concluding section will comment on the significance of the results.

Fi

Fig.

Some articles present data on a series of rare-earth ions replacing La in LaSrCuO or Y in YBaCuO and others report only particular rare-earth-substituted compounds. Several papers involve members of the first transition series and other atoms replacing Cu, and some substitutions for O will also be presented. The magnetic behavior of the rare earths substituted in YBaCuO will be discussed in Section VIII-D-3.

#### 1. Rare Earths in LaSrCuO

Several authors substituted Nd (namely, Crab1, Kwok2) and various rare-earth elements R for lanthanum in  $(La_{1-x-y}R_ySr_x)_2CuO_{4-\delta}$  (namely, Dikoz, Fueki, Grove, Haseg, Hoso1, Kish1, McKin, Ogita, Phata, Tara5), and this causes a lowering of the transition temperature, as the normalized resistivity plots of Fig. VII-8 demonstrate. Room-temperature resistivities for this series of compounds varied between 1600 and 2540  $\mu\Omega$  cm, and the ratio  $p_{300K}/p_{50K}$  varied between 2.9 and 4.6. In some cases double substitutions such as Ba<sub>x</sub>Sr<sub>y-x</sub> for lanthanum were employed (e.g., Hosoy). Figure VII-9 shows how  $T_c$  varies across the rare-earth series.

#### 2. Rare Earths in YBaCuO

Many investigators have studied rare earth substitutions in the YBaCuO system (namely, Chuz2, Fiskz, Horzz, Kurih, Kuzzz, Lepa1, Maple, Marcu, McKin,

conclusive (cf. Section

ited to perovskite fermaterials were aided transitions in oxides ide a channel for the

O and YBaCuO each Sr, Ba), copper, and of the constituent aterhaps not so related oth  $(R_{1-x}M_x)_2CuO_{4-\delta}$  with copper accounttutions and total rescluding section will

ons replacing La in ar rare-earth-substiirst transition series · O will also be pre-1 in YBaCuO will be

d various rare-earth rely, Dikoz, Fueki, ), and this causes a sistivity plots of Fig. reries of compounds  $o_{SOK}$  varied between  $Sr_{y-x}$  for lanthanum ries across the rare-

ne YBaCuO system e, Marcu, McKin,

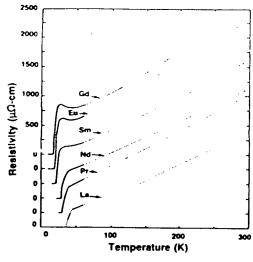


Fig. VII-8. Resistivity versus temperature for rare-earth (R) substitutions in the compound  $(La_{0.8}Sr_{0.1}R_{0.1})_2$  CuO<sub>4- $\delta$ </sub> (Tara5).

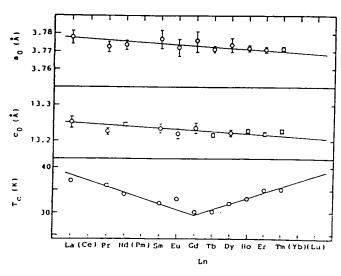


Fig. VII-9. Variations of tetragonal lattice parameters  $a_0$  and  $c_0$  and ac susceptibility onset  $T_c$  for the various rare earth ion (R) substitutions (La<sub>0.85</sub>Sr<sub>0.1</sub>R<sub>0.05</sub>)<sub>2</sub>CuO<sub>4-6</sub> (Kish1).

Murph, Namzz, Parki, Tanal, Tara4, Torra, Xiaol, Yamad, Yangz, Yangl, Yang2), and the subject has been reviewed (Tara2). Experimental data on particular rare earths replacing Y in YBaCuO have been reported in individual articles, and some examples are dysprosium (Qiru1), erbium (Ayyub, Golbe, Hayri, Qiru3, Zuozz), europium (Boolc), gadolinium (Escu2, Podda, Qiru2, Thomp, Zuozz), holmium (Hayri, Kagol, Leez2, Podda, Thom2, Zuozz), lanthanum (Cheva, Leez3, Segre), lutetium (Raych), neodymium (Escu2), praseodymium (Dalic, Soder), scandium (Shizz, Zhao1), samarium (Csach, Garci, Hajko), thulium (Andr1, Neume), and ytterbium (Grove, Qiru3). This list of individual elements does not include articles that involve several rare-earth substitutions.

Typically all of the yttrium was replaced by a rare-earth ion, and this total replacement produced lattice constant changes (Eagle, Horzz, Tara4, Yang2), as shown on Fig. VII-10. The results summarized in Table VII-2 demonstrate that most of the rare earths produce transition temperatures of 85-95 K, and several of them (Eu, Dy, Ho, Tm, and Yb) achieved narrow transitions with  $T_c$  above 90 K. Four rare-earth elements (Ce, Pr, Pm, and Tb) did not form superconductors. Also included in the table are values of the resistivity at 300 K and at the temperature of the onset of superconductivity together with lattice constants and susceptibility data. Figure VII-11 presents a typical resistivity versus temperature plot for various substitutions (Maple, McKin, Murph, Tara2-Tara5, Yangz).

#### 3. Alkaline Earths

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Many measurements have been made on barium-substituted and some on calcium-substituted La<sub>2</sub>CuO<sub>4- $\delta$ </sub> (Hosoy, Terak), and data on these compounds are found in several places throughout this review. The Sr<sup>2+</sup> ion, which has a size (1.12 Å) close to that of La<sup>3+</sup> (1.14 Å), produces higher transition temperatures and three times the diamagnetic susceptibility in  $(La_{1-x}M_x)_2CuO_{4-\delta}$  than the larger Ba<sup>2+</sup> (1.34 Å) ion (Bedn2).

When strontium is substituted for barium in the system  $Y(Ba_{1-x}Sr_x)_2Cu_3O_{7-\delta}$ , the transition temperature drops from 90 K to about 78 K in the range from x=0 to 0.75 (Qirui, Wuzz2). Meissner flux exclusion is about 45% from x=0 to x=0.5 and drops off thereafter (Vealz). Therefore, the x=0 composition is the best.

#### 4. Paramagnetic Substitutions for Copper

The series of compounds YBa<sub>2</sub>(Cu<sub>0.9</sub>M<sub>0.1</sub>)<sub>3</sub>O<sub>7- $\delta$ </sub> where M is a member of the first transition series of elements was fabricated with the oxygen deficiency parameter  $\delta$  undetermined (Xiaoz). All of the transition elements reduced  $T_c$ , but to a different extent, as shown on the resistivity plots of Fig. VII-12. The susceptibility  $\chi$  above  $T_c$  is described by a temperature-independent part  $\chi_0$  and a paramagnetic part, as is explained in Section VIII-D-1. The depression of  $T_c$  correlates with the magnetic moment of the substituted transition ion, the larger the mo-

Yamad, Yangz, Yangl, xperimental data on pareported in individual artism (Ayyub, Golbe, Hayri, Podda, Qiru2, Thomp, m2, Zuozz), lanthanum (Escu2), praseodymium sach, Garci, Hajko), thu-This list of individual eleve-earth substitutions.

-earth ion, and this total, Horzz, Tara4, Yang2), Table VII-2 demonstrate ratures of 85-95 K, and arrow transitions with  $T_c$  i Tb) did not form superresistivity at 300 K and at her with lattice constants cal resistivity versus temin, Murph, Tara2-Tara5,

tituted and some on calon these compounds are  $r^{2+}$  ion, which has a size r transition temperatures  $a_{1-x}M_x)_2CuO_{4-\delta}$  than the

tem Y(Ba<sub>1-x</sub>Sr<sub>x</sub>)<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>, K in the range from x = bout 45% from x = 0 to x = 0 composition is the

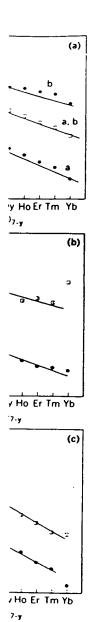
I is a member of the first tygen deficiency parametrs reduced  $T_c$ , but to a VII-12. The susceptibility part  $\chi_0$  and a paramagripression of  $T_c$  correlates n ion, the larger the mo-

TABLE VII.2. Effect of Substituting Various Atoms in R and M Sites of RM<sub>2</sub>Cu<sub>3</sub>O<sub>7.3</sub>"

11.67   260   2.3   10.9     11.70	~	Σ	7; (K)	Δ7'(K)	" (Ý)	/ <sub>(</sub> , (Å)	c (Å)	ρ <sub>onet</sub> μΩ·cm	P.3001/Primer	κοχ	χωε/χ <sub>DS</sub> (%)	Ref.
Ba 59.2 17.9 3.94 3.95 11.97 — — — — — — — — — — — — — — — — — — —	<b>~</b>	Ва	93.3	2.1	3.86	3.87	11.67	260	2.3	10 0	5	V
Ba         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —	La	Ва	59.2	17.9	3.94	3.95	11.97	; ;	<u></u>	<u>:</u>	10	* 0 :: * * *
Ba         —         3.886         3.912         11.710         —         3.1           Ba         78.3         28         3.882         3.96         11.746         —         3.1           Ba         78.3         28         3.882         3.96         11.74         —         —         3.1           Ba         88.6         11.9         3.83         3.89         11.75         740         2.5         14.2           Ba         90.9         7.4         3.845         11.73         —         —         —         19.1           Ba         90.9         7.4         3.845         3.89         11.73         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —         —	ల	Ba	ı	1	i	}	:		l	I	í	W.B.
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90.5       1.7       3.840       3.883       11.574       —       —       13.9         72.6       1.3       3.83       3.84       11.63       —       —       10.7         91       5       3.84       3.86       11.74       1860       2.15       —         90       4       —       —       —       —       5.7         90       4       —       —       —       —       5.7         90       4       —       —       —       —       5.7         90       4       —       —       —       —       —       5.7         90       4       —       —       —       —       —       —       —       —       5.7         90       4       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —       —	ŗ.	Ba	7.06	4.9	3.838	3.862	11.586	ł	ı	12.8	4	Ave.
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The rows give from left to right the R and M atoms; the transition temperature Te and its width DT; the three lattice parameters a, b, c; resistivity data for the onset. powers the ratio with room-temperature value pina; and the diamagnetic shielding (DS) and Meissner (ME) susceptibility. The rows with an asterisk (4) contain transition temperatures Te. A T and lattice parameters a.b.c averaged from Engle, Horzz, Kurih, Kuzzz, Maple, McKin, Tara3, Yamad, Yangz, Kish2.

Fig. VII-10. Orthorhombic unit-cell parameters (a,b,c) and unit-cell volume (V) as a function of the rare earth R in the RBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> series for the as-prepared samples ( $\blacksquare$ ) and vacuum-annealed samples ( $\square$ ) are shown. The lines drawn are a guide for the eye (Tara4).



) and unit-cell volume (V) as a or the as-prepared samples (lacktriangle) s drawn are a guide for the eye

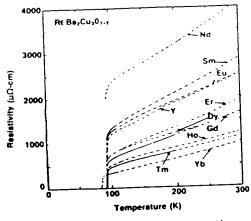


Fig. VII-11. Resistivity as a function of temperature for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub> and compounds for which a rare earth has been substituted for the Y (Tara2).

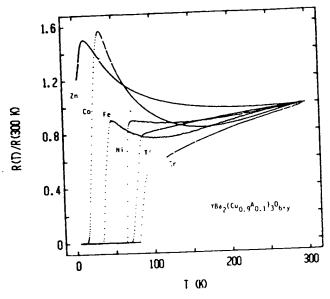


Fig. VII-12. Temperature dependence of the normalized resistance of  $Y^{p_-}$ .  $(Cu_{0.9}A_{0.1})_3O_{7-\delta}$ , where A = Ti, Cr, Fe, Co, Ni, and Zn (From Xiaoz).

ment, the lower the  $T_c$  value, as shown by the data in Fig. VII-13. Comparing with Fig. II-2, we see that there are two maxima for  $T_c$ , but they do not occur for the same number of valence electrons. The Cu maximum at  $N_c = 11$  on Fig. VII-13 is beyond the region of the curves shown on Fig. II-2. Others (Felne, CuO (Haseg)).

An extensive examination was made of the effect of substituting paramagnetic Ni<sup>2+</sup> and diamagnetic Zn<sup>2+</sup> ions (M) for copper ions in (La<sub>0.925</sub>Sr<sub>0.075</sub>)<sub>2</sub>-Cu<sub>1-x</sub>M<sub>x</sub>O<sub>4</sub> and YBa<sub>2</sub>(Cu<sub>1-x</sub>M<sub>x</sub>)<sub>3</sub>O<sub>7-\delta</sub> (Csach, Tara3, Tara6, Tara8, Thiel). Changes in lattice constants, transition temperature, susceptibility, and upper critical fields were determined. Variations in unit-cell dimensions may result YBaCuO system degrades the superconductivity to a much lesser extent than in the LaSrCuO case (Tara3, Tara8). This lessened effect relative to LaSrCuO may cu-O layers where the Cooper pairs reside. The Cu-O layers in the LaSrCuO compound, on the other hand, are all equivalent and contribute equally to the superconductivity.

### 5. Nonmagnetic Substitutions for Copper

Zinc and Ga have definite valence states of +2 and +3 and ionic radii 0.74 and 0.62, respectively (cf. Table VI-2), which reflect those of  $Cu^{2+}$  and  $Cu^{3+}$ . There-

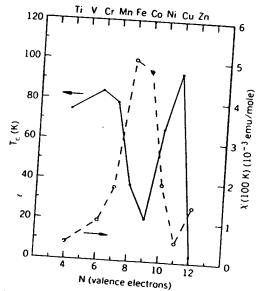


Fig. VII-13. Transition temperature (——) and magnetic susceptibility at 100 K (---) of YBa<sub>2</sub>(Cu<sub>0.9</sub>M<sub>0.1</sub>)<sub>3</sub>Q<sub>7-6</sub>, where M is a 3d transition element, as a function of the number of valence electrons (from Xiaoz).

VII-13. Comparing t they do not occur at  $N_e = 11$  on Fig. 1-2. Others (Felne, and also for LaSr-

stituting paramagin (La<sub>0.925</sub>Sr<sub>0.075</sub>)<sub>2</sub>-6, Tara8, Thiel). tibility, and upper ensions may result itution of Ni in the esser extent than in e to LaSrCuO may t on the particulars in the LaSrCuO oute equally to the

nic radii 0.74 and and Cu<sup>3+</sup>. There-

fore Zn should replace  $Cu^{2+}$  on the  $Cu-O_2$  planes, and 15% replacement is enough to completely destroy the superconductivity. In like manner Ga should substitute for  $Cu^{3+}$  on the Cu-O chain layer, and substitutions up to 20% reduce  $T_c$  by only 30%. In addition gallium contents above 4% induce the tetragonal-to-orthorhombic transformation (Xiao3).

The substitution of Ag. which is ordinarily monovalent, for Cu in  $(La_{0.9} Sr_{0.1})_2Cu_{1-x}Ag_xO_{4-\delta}$  depressed  $T_c$  by 50% at the x=0.15 level (Malik), much less than the effect of Ni or Zn.

Partial replacement of Cu by monovalent Ag in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> depressed  $T_c$  (Tomyz), and in one report total replacement lowered the onset  $T_c$  to 50 K and increased the transition width to 30 K (Panz1). This replacement increased the Cu<sup>3+</sup>/Cu<sup>2+</sup> ratio, presumably for charge balance, although an increase in oxygen vacancies may also occur. Ag replacement was found to enhance the critical current density by a factor of 15 (Kungz).

#### 6. Substitutions for Oxygen

Several studies have been made with fluorine replacing part of the oxygen (Ovshi, Tonou). The structure of  $YBa_2Cu_3O_6F_2$  may be tetragonal with fluorines occupying all of the O(t) and O(t') sites on the z=0 basal plane and oxygen occupying all of its other sites (cf. Table VI-6). This arrangement would eliminate oxygen site vacancies and chains, with all of the copper ions in square-planar nearest-neighbor coordination. Implanted fluorine and its diffusion were also studied (Kiste, Tesme, Xianr).

Zero resistance has been claimed at  $T_c = 155 \text{ K}$  for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6</sub>F<sub>2</sub> (Ovshi) and at 148.5 K with fluorine implantation (Xianr).

The replacement of part of the oxygen by sulphur can form the compound  $YBa_2Cu_3O_6S$ , which has a sharper transition without changing  $T_c$ . The compound displays the full Meissner effect (Felne). Total oxygen replacement has not (yet) been achieved, but partial replacement by either F or S is not necessarily destructive and can enhance the superconducting properties (Feln1). Sulphur can also transform the crystal to the tetragonal form.

#### 7. Other Substitutions

Aluminum substitution in  $Y_{1-x}AI_xBa_2Cu_3O_{7-\delta}$  for 0 < x < 0.85 (Franc) and in BiSrCaCuO (Chuz5) lowers  $T_c$ . Another report showed that small quantities of aluminum ( $x = 0.05, 0.08, y \approx 0.35$ ) in the system  $(Y_{1-x-y}Ba_xAI_y)_2CuO_4$  do not effect the onset  $T_c$  (Escu1).

#### 8. Influence of Substitutions on Superconductivity

We have seen that substitutions of magnetic ions for La or Y have very little effect on the superconductivity while, in general, substitutions for Cu have a

y at 100 K (---) of n of the number of

60

destructive effect. This supports the belief that the superconducting quasi-particles or Cooper pairs are associated with the Cu-O layers, and that the La, Ba, and Y layers are not directly involved in the superconductivity mechanism.

In ordinary or low-temperature superconductors the presence of magnetic ions destroys the superconductivity quite strongly (Matt1), while in the LaSrCuO and YBaCuO cases the destruction is selective, depending on how close the substitutions are to the Cooper-pair layers (Haseg, Hoso1).

It is doubtless significant that a high  $T_c$  superconducting compound can be obtained with all of the Cu replaced by Ag and with much of it replaced by Ga. It is also significant that F and S substitutions for O are not necessarily deleterious.

#### G. ISOTOPE EFFECT

The traditional test for phonon-mediated superconductivity is the isotope effect (Section III-G-2), whereby for an element  $T_{\rm c}$  is related to the atomic mass M through the expression

$$T_{\rm c} {\rm M}^{\alpha} = {\rm const}$$
 (VII-4)

with the isotope effect coefficient  $\alpha = \frac{1}{2}$  for electron-phonon coupling in elemental superconductors. For compounds with many atoms it is difficult to predict how  $T_c$  depends on the mass of a particular isotope in a complex structure, but isotope effects are observed in compounds. Several investigators report that <sup>18</sup>O substitution on the two sites of LaSrCuO produce the positive isotope effect  $0.1 < \alpha < 0.35$ , as shown in Fig. VII-14 (Batl1, Batl2, Cohe1, Falte). In contrast, the results for YBaCuO have been partly negative. The replacement of 10O by <sup>18</sup>O in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> and EuBa<sub>2</sub>CuO<sub>7- $\delta$ </sub> gave  $\alpha \approx 0$  (Batlo, Batl2, Bourn, Cohe1, Morri). The absence of an isotope effect with enriched 63Cu, 65Cu, 135Ba, and 138Ba was also reported (Bour4). Nevertheless, there were reports (Katay, Leary) of a decrease in  $T_c$  of 0.2-0.5 K by the 70-90% replacement of  $^{16}O$  by  $^{18}O$ . as indicated in Fig. VII-15. The same magnitude of shift was found for the four oxide superconductors  $BaBi_{0.25}Pb_{0.75}$  with  $T_c = 11$  K,  $(La_{0.925}Ca_{.075})_2CuO_4$  with  $T_c = 20 \text{ K}$ ,  $(La_{0.925}Sr_{.075})_2CuO_4$  with  $T_c = 37 \text{ K}$ , and  $YBa_2Cu_3O_7$  with  $T_c = 92 \text{ K}$ . with a small dependence on  $T_c$  (Stacy). In addition, Raman phonon frequencies do exhibit the expected 4% mass dependent shifts (Batlo).

A positive result of  $\alpha$  close to  $\frac{1}{2}$  is strong support for a phonon-mediated BCS mechanism, as was noted in Section IV-B-2. The isotope effect coefficient increases slowly with coupling strength (Mars2). Some transition ion superconductors such as Os and Nb<sub>3</sub>Sn have unusually small isotopic mass dependencies, and for others such as Ru and Zr,  $T_c$  is independent of M (Table II-1), so negative results do not preclude the operation of a phonon mechanism. Also, other mechanisms such as excitons, resonant valence bonds, and the librational model of e-e pairing (Hardy) are predicted to exhibit little or no isotopic shift.

#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J. Bednorz et al. Date: December 15, 1998

Serial No. 08/303,561 Group Art Unit: 1105

Filed: September 9, 1994 Examiner: M. Kopec

For: NEW SUPERCONDUCTIVE COMPOUNDS HAVING HIGH TRANSITION TEMPERATURE, AND METHODS FOR THEIR

**USE AND PREPARATION** 

The Commissioner of Patents and Trademarks Washington, D.C. 20231

#### **AFFIDAVIT UNDER 37 CFR 1.132**

Sir:

I, James W. Leonard, being duly sworn, do hereby depose and state:

I received a A.B. degree in Physics from the University of California at Santa Barbara (1962), and a M.S. and PhD. degree in Physics (1968) from the University of Oregon, Eugene, and an M.L.S. in Library Science from the University of Western Ontario (1972), London.

I have worked as a science librarian in the Thomas J. Watson Research Center from 1978 to the present. On December 2, 1998, I did a citation search in the SciSearch database on the Dialog on line system of the article J. G. Bednorz and K. A. Muller, Zeitschrift fur Physik B- Condensed Matter, 64, pp. 189-193 (Sept. 1986). The result

of that search is below. There are a total of 5689 articles which refer to 1986 article of Bednorz and Muller: 1 in 1986, 839 in 1987, 1163 in 1988, 793 in 1989, 594 in 1989 and the remainder in the years from 1990 to the present.

#### **SEARCH RESULTS**

SYSTEM:OS - DIALOG OneSearch
File 434:SciSearch(R) Cited Ref Sci 1974-1989/Dec
(c) 1998 Inst for Sci Info
File 34:SciSearch(R) Cited Ref Sci 1990-1998/Nov W4
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Set	Items	Description
S1	5689	CR=BEDNORZ JG, 1986, V64, P189, ?
S2	1	S1 AND PY=1986
S3	839	S1 AND PY=1987
S4	1163	S1 AND PY=1988
S5	793	S1 AND PY=1989
S6	594	S1 AND PY=1990

By: James W. Leonard

Sworn to before me this

day of

19 98

Notary Public

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Commission Expires March 16, 19-27

# COPPER OXIDE SUPERCONDUCTORS

Charles P. Poole, Jr. Timir Datta Horacio A. Farach

with help from

M. M. Rigney C. R. Sanders

Department of Physics and Astronomy University of South Carolina Columbia, South Carolina



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#### **PREFACE**

The unprecedented worldwide effort in superconductivity research-that has taken place over the past two years has produced an enormous amount of experimental data on the properties of the copper oxide type materials that exhibit superconductivity above the temperature of liquid nitrogen. The time is now ripe to bring together in one place the results of this research effort so that scientists working in this field can better acquire an overall perspective, and at the same time have available in one place a collection of detailed experimental data. This volume reviews the experimental aspects of the field of oxide superconductivity with transition temperatures from 30 K to above 120 K, from the time of its discovery by Bednorz and Müller in April 1986 until a few months after the award of the Nobel Prize to them in October 1987. During this period a consistent experimental description of many of the properties of the principal superconducting compounds such as BiSrCaCuO, LaSrCuO, TIBaCaCuO, and YBa-CuO has emerged. At the same time there has been a continual debate on the extent to which the BCS theory and the electron-phonon interaction mechanism apply to the new materials, and new theoretical models are periodically proposed. We discuss these matters and, when appropriate, make comparisons with transition metal and other previously known superconductors. Many of the experimental results are summarized in figures and tables.

The field of high-temperature superconductivity is still evolving, and some ideas and explanations may be changed by the time these notes appear in print. Nevertheless, it is helpful to discuss them here to give insights into work now in progress, to give coherence to the present work, and to provide guidance for future work. It is hoped that in the not too distant future the field will settle

down enough to permit a more definitive monograph to be written.

#### vi PREFACE

The literature has been covered almost to the end of 1987, and some 1988 work has been discussed. This has been an enormous task, and we apologize for any omissions in the citing and discussion of articles.

We wish to thank the following for giving us some advanced notice about their work: R. Barrio, B. Battlogg, L. A. Boatener, G. Burns, J. Drumheller, H. Enomoto, P. K. Gallagher, R. Goldfarb, J. E. Graebner, R. L. Greene, J. Heremans, T. C. Johnson, J. K. Karamas, M. Levy, J. W. Lynn, A. Malozemoff, K. A. Müller, T. Nishino, N. Nucker, J. C. Phillips, R. M. Silver, G. Shirane, J. Stankowski, B. Stridzker, S. Tanigawa, G. A. Thomas, and W. H. Weber. We appreciate comments on the manuscript from S. Alterowitz, C. L. Chien, D. K. Finnamore, J. Goodenough, J. R. Morton, and C. Uher, and helpful discussions with J. Budnick, M. H. Cohen, M. L. Cohen, R. Creswick, S. Deb, M. Fluss, A. Freeman, D. U. Gubser, A. M. Hermann, V. Z. Kresin, H. Ledbetter, W. E. Pickett, M. Tinkham, C. E. Violet, and S. A. Wolf. Support from the University of South Carolina, the Naval Research Laboratory, and the National Science Foundation Grant ISP 80 11451 is gratefully acknowledged.

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Charles P. Poole, Jr. Timir Datta Horacio A. Farach

Columbia, South Carolina July 1988 ects of the BCS theory, however,

id detailed treatment of the propsee the extent to which they coney agree with some of the other in these two chapters.



### PREPARATION AND CHARACTERIZATION OF SAMPLES

#### A. INTRODUCTION

Copper oxide superconductors with a purity sufficient to exhibit zero resistivity or to demonstrate levitation (Early) are not difficult to synthesize. We believe that this is at least partially responsible for the explosive worldwide growth in these materials. Nevertheless, it should be emphasized that the preparation of these samples does involve some risks since the procedures are carried out at quite high temperatures, often in oxygen atmospheres. In addition, some of the chemicals are toxic, and in the case of thallium compounds the degree of toxicity is extremely high so ingestion, inhalation, and contact with the skin must be prevented.

The superconducting properties of the copper oxide compounds are quite sensitive to the method of preparation and annealing. Multiphase samples containing fractions with  $T_{\rm c}$  above liquid nitrogen temperature (Monec) can be synthesized using rather crude techniques, but really high-grade single-phase specimens require careful attention to such factors as temperature control, oxygen content of the surrounding gas, annealing cycles, grain sizes, and pelletizing procedures. The ratio of cations in the final sample is important, but even more critical and more difficult to control is the oxygen content. However, in the case of the Bi- and Tl-based compounds, the superconducting properties are less sensitive to the oxygen content.

Figure V-1 illustrates how preparation conditions can influence superconducting properties. It shows how the calcination temperature, the annealing time, and the quenching conditions affect the resistivity drop at  $T_c$  of a BiSrCa-CuO pellet, a related copper-enriched specimen, and an aluminum-doped coun-



#### PREPARATION AND CHARACTERIZATION OF SAMPLES

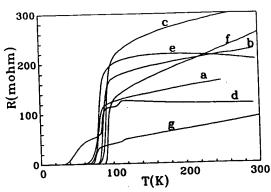


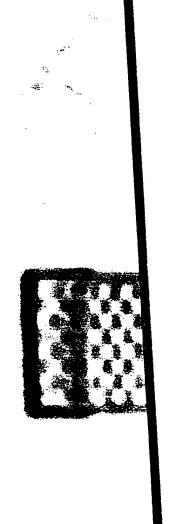
Fig. V-1. Effects of heat treatments on the resistivity transition of BiSrCaCuO<sub>1,4</sub>(a) calcined at 860°C, (b) calcined at 885°C, (c) calcined at 901°C, (d) aluminum-doped sample calcined at 875°C, prolonged annealing, (e) copper-rich sample calcined at 860°C, (f) aluminum-doped sample calcined at 885°C, slow quenching and (g) calcined at 885°C, prolonged annealing, and slow quenching (Chuz5).

terpart (Chuz5). These samples were all calcined and annealed in the same temperature range and air-quenched to room temperature.

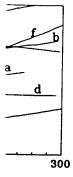
Polycrystalline samples are the easiest to prepare, and much of the early work was carried out with them. Of greater significance is work carried out with this films and single crystals, and these require more specialized preparation techniques. More and more of the recent work has been done with such samples.

Many authors have provided sample preparation information, and other have detailed heat treatments and oxygen control. Some representative techniques will be discussed.

The beginning of this chapter will treat methods of preparing bulk superconducting samples in general, and then samples of special types such as thin films and single crystals. The remainder of the chapter will discuss ways of checking the composition and quality of the samples. The thermodynamic or subsolidate phase diagram of the ternary Y-Ba-Cu oxide system illustrated in Fig. V-2 contains several stable stoichiometric compounds such as the end-point oxides Y<sub>2</sub>O<sub>3</sub>, BaO, and CuO at the apices, the binary oxides stable at 950°, (Ba<sub>3</sub>CuO<sub>3</sub>, Ba<sub>2</sub>CuO<sub>3</sub>, BaCuO<sub>2</sub>, Y<sub>2</sub>Cu<sub>2</sub>O<sub>5</sub>, Y<sub>4</sub>Ba<sub>3</sub>O<sub>9</sub>, Y<sub>2</sub>BaO<sub>4</sub>, and (Y<sub>2</sub>Ba<sub>4</sub>O<sub>7</sub>), along the edges, and ternary oxides such as (YBa<sub>3</sub>Cu<sub>2</sub>O<sub>7</sub>), the semiconducting green phase Y<sub>2</sub>BaCuO<sub>5</sub>, and the superconducting black solid YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub> in the interior (Beye2, Bour3, Capo1, Eagl1, Frase, Hosoy, Jone1, Kaise, Kurth, Kuzza, Leez3, Lian1, Mali1, Schni, Schn1, Schu1, Takay, Torra, Wagne). Compounds in parentheses are not on the figure, but are reported by other workers. The existence of a narrow range of solid solution was reported (Panso), and then argued against (Wagne) by the same group.



MPLES



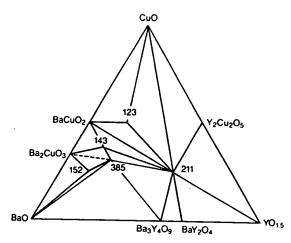
cansition of BiSrCaCuO<sub>7-8</sub> (a) t 901°C, (d) aluminum-doped pper-rich sample calcined at ow quenching and (g) calcined 125).

annealed in the same tem-

and much of the early work work carried out with thin cialized preparation techdone with such samples. In information, and others

Some representative tech-

preparing bulk superconial types such as thin films discuss ways of checking modynamic or subsolidus llustrated in Fig. V-2 conas the end-point oxides stable at 950°, (Ba<sub>3</sub>CuO<sub>4</sub>), nd (Y<sub>2</sub>Ba<sub>4</sub>O<sub>7</sub>), along the miconducting green phase la<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub> in the interior l, Kaise, Kurth, Kuzzz, rra, Wagne). Compounds d by other workers. The ported (Panso), and then



Compound .	Slowly cooled to room temperature
123 - YBa <sub>2</sub> Cu <sub>3</sub> O <sub>6.5+ è</sub>	07
143 - YBa₄Cu <sub>3</sub> O <sub>8.5+ δ</sub>	O <sub>9</sub>
385 - Y3Ba8Cu5O17.5+ 8	O <sub>18</sub>
152 - YBa <sub>5</sub> Cu <sub>2</sub> O <sub>8.5+ ծ</sub>	O <sub>9</sub>
211 - Y <sub>2</sub> BaCuO <sub>5</sub>	·
Ba <sub>2</sub> CuO <sub>3+&amp;</sub>	O <sub>33</sub>

Fig. V-2. Ternary phase diagram of the  $Y_2O_3$ -BaO-CuO system at 950°C. The green phase  $[Y_2BaCuO_5, (211)]$  the superconducting phase  $[YBa_2Cu_3O_{7-\delta}, (123)]$ , and three other compounds are shown in the interior of the diagram (DeLee).

#### **B. METHODS OF PREPARATION**

In this section three methods of preparation will be described, namely, the solid state, the coprecipitation, and the sol-gel techniques (Hatfi). The widely used solid-state technique permits off-the-shelf chemicals to be directly calcined into superconductors, and it requires little familiarity with the subtle physicochemical processes involved in the transformation of a mixture of compounds into a superconductor. The coprecipitation technique mixes the constituents on an atomic scale and forms fine powders, but it requires careful control of the pH and some familiarity with analytical chemistry. The sol-gel procedure requires more competence in analytical procedures.

In the solid-state reaction technique one starts with oxygen-rich compounds of the desired components such as oxides, nitrates, or carbonates of Ba, Bi, La, Sr, Tl, Y, or other elements. Sometimes nitrates are formed first by dissolving oxides in nitric acid and decomposing the solution at 500°C before calcination

(e.g., Davis, Holla, Kelle). These compounds are mixed in the desired atomic ratios and ground to a fine powder to facilitate the calcination process. Then these room-temperature-stable salts are reacted by calcining for an extended period (≈ 20 hr) at elevated temperatures (≈ 900°C). This process may be repeated several times, with pulverizing and mixing of the partially calcined material at each step. As the reaction proceeds, the color of the charge changes. The process usually ends with a final oxygen anneal followed by a slow cool down to room temperature of the powder, or pellets made from the powder, by sintering in a cold or hot press. Sintering is not essential for the chemical process, but for transport and other measurements it is convenient to have the material pelletized. A number of researchers have provided information on this solid-state reaction approach (e.g., Allge, Finez, Galla, Garla, Gopal, Gubse, Hajk1, Hatan, Herrm, Hika1, Hirab, Jayar, Maen1, Mood1, Mood2, Neume, Poepp, Polle, Qadri, Rhyne, Ruzic, Saito, Sait1, Sawa1, Shamo, Takit, Tothz, Wuzz3).

Some of the earlier works on foils, thick films, wires, or coatings employed a suspension of the calcined powder in a suitable organic binder, and the desired product was obtained by conventional industrial processes such as extruding, spraying, or coating.

In the second or coprecipitation process the starting materials for calcination are produced by precipitating them together from solution (e.g., Asela, Bedno, Leez7, Wang2). This has the advantage of mixing the constituents on an atomic scale. In addition the precipitates may form fine powders whose uniformity can be controlled, which can eliminate some of the labor. Once the precipitate has been dried, calcining can begin as in the solid-state reaction procedure. A disadvantage of this method, at least as far as the average physicist or materials scientist is concerned, is that it requires considerable skill in chemical procedures.

Another procedure for obtaining the start-up powder is the sol-gel technique in which an aqueous solution containing the proper ratios of Ba, Cu, and Y nitrates is emulsified in an organic phase and the resulting droplets are gelled by the addition of a high-molecular-weight primary amine which extracts the nitric acid. This process was initially applied to the La materials, but has been perfected for YBaCuO as well (Cimaz, Hatfi).

When using commercial chemical supplies to facilitate the calcination process a dry or wet (acetone) pregrinding with an agate mortar and pestle or a ball mill is recommended. Gravimetric amounts of the powdered precursor materials are thoroughly mixed and placed in a platinum or ceramic crucible. Care must be taken to ensure the compatibility of the ceramic crucible with the chemicals to obviate reaction and corrosion problems.

Complete recipes for the YBa\* material have been described (e.g., Gran2). Typically, the mixture of unreacted oxides is calcined in air or oxygen around 900°C for 15 hr. During this time the YBaCuO mixture changes color from the green Y<sub>2</sub>BaCuO<sub>5</sub> phase to the dark gray YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub> compound. Then the charge is taken out, crushed, and scanned with X rays to determine its purity. If warranted by the powder pattern X-ray scan, the calcination process is repeated. Often, at this stage the material is very oxygen poor, and electrically it is semi-

Attachment H

# COPPER OXIDE SUPERCONDUCTORS

Charles P. Poole, Jr. Timir Datta Horacio A. Farach

with help from

M. M. Rigney C. R. Sanders

Department of Physics and Astronomy University of South Carolina Columbia, South Carolina



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Printed in the United States of America

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#### **PREFACE**

The unprecedented worldwide effort in superconductivity research that has taken place over the past two years has produced an enormous amount of experimental data on the properties of the copper oxide type materials that exhibit superconductivity above the temperature of liquid nitrogen. The time is now ripe to bring together in one place the results of this research effort so that scientists working in this field can better acquire an overall perspective, and at the same time have available in one place a collection of detailed experimental data. This volume reviews the experimental aspects of the field of oxide superconductivity with transition temperatures from 30 K to above 120 K, from the time of its discovery by Bednorz and Müller in April 1986 until a few months after the award of the Nobel Prize to them in October 1987. During this period a consistent experimental description of many of the properties of the principal superconducting compounds such as BiSrCaCuO, LaSrCuO, TlBaCaCuO, and YBa-CuO has emerged. At the same time there has been a continual debate on the extent to which the BCS theory and the electron-phonon interaction mechanism apply to the new materials, and new theoretical models are periodically proposed. We discuss these matters and, when appropriate, make comparisons with transition metal and other previously known superconductors. Many of the experimental results are summarized in figures and tables.

The field of high-temperature superconductivity is still evolving, and some ideas and explanations may be changed by the time these notes appear in print. Nevertheless, it is helpful to discuss them here to give insights into work now in progress, to give coherence to the present work, and to provide guidance for future work. It is hoped that in the not too distant future the field will settle down enough to permit a more definitive monograph to be written.

The literature has been covered almost to the end of 1987, and some 1988 work has been discussed. This has been an enormous task, and we apologize for any omissions in the citing and discussion of articles.

We wish to thank the following for giving us some advanced notice about their work: R. Barrio, B. Battlogg, L. A. Boatener, G. Burns, J. Drumheller, H. Enomoto, P. K. Gallagher, R. Goldfarb, J. E. Graebner, R. L. Greene, J. Heremans, T. C. Johnson, J. K. Karamas, M. Levy, J. W. Lynn, A. Malozemoff, K. A. Müller, T. Nishino, N. Nucker, J. C. Phillips, R. M. Silver, G. Shirane, J. Stankowski, B. Stridzker, S. Tanigawa, G. A. Thomas, and W. H. Weber. We appreciate comments on the manuscript from S. Alterowitz, C. L. Chien, D. K. Finnamore, J. Goodenough, J. R. Morton, and C. Uher, and helpful discussions with J. Budnick, M. H. Cohen, M. L. Cohen, R. Creswick, S. Deb, M. Fluss, A. Freeman, D. U. Gubser, A. M. Hermann, V. Z. Kresin, H. Ledbetter, W. E. Pickett, M. Tinkham, C. E. Violet, and S. A. Wolf. Support from the University of South Carolina, the Naval Research Laboratory, and the National Science Foundation Grant ISP 80 11451 is gratefully acknowledged.

Michael A. Poole helped to develop the computer data storage techniques that were used. Jesse S. Cook is thanked for editorial comments on the manuscript. C. Almasan, S. Atkas, J. Estrada, N. Hong, O. Lopez, M. Mesa, T. Mouzghi, and T. Usher are thanked for their interest in this project.

Charles P. Poole, Jr. Timir Datta Horacio A. Farach

Columbia, South Carolina July 1988 Attachment A

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<del>\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*</del>06068<del>\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*</del> LEVEL 1 - 1 OF 68 PATENTS

5,670,078

<=2> GET 1st DRAWING SHEET OF 7

Sep. 23, 1997

Magnetic and nonmagnetic particles and fluid, methods of making and methods of using the same

INVENTOR: Ziolo, Ronald F., Webster, New York

**DETDESC:** 

... described in U.S. Pat. No. 4,474,886 to Ziolo. Examples of the precursor ions which may be used includes those derivable from transition metal ions, such as iron, cobalt, nickel, manganese, vanadium, chromium, rare earths and the like. In the case of a mon-magnetic colloid, this may include ions of, for example, sulfur, selenium, gold, barium, cadmium, copper, silver, manganese, molybdenum, zirconium; gallium; arsonic, indium, tin, ... الرواي الماليكيك وسيتحب يه

... ions which can be incorporated into the resin beads to form both

single-domain and multidomain magnetic particles include: those derivable from transition metal ions, such as iron, cobalt, nickel, manganese, vanadium, chromium, rare earths and the like. These ions generally exist in the form of chlorides of the metal involved such as ferrous chloride, ferric chloride, copper chloride, nickel chloride, and the like. The corresponding iodides, bromides and fluorides may also be suitable. ...

LEVEL 1 - 2 OF 68 PATENTS

5,663,319

Sep. 2, 1997

Probe compositions for chromosome identification and methods

INVENTOR: Bittner, Michael L., Naperville, Illinois Morrison, Larry E., DuPage County, Illinois Legator, Mona S., Chicago, Illinois

#### SUM:

... capable of reacting, and a fluorophore group may have already reacted, with a linking group. A fluorescent compound may include an organic chelator which binds a luminescent inorganic ion such as a rare earth like terbium, europium, ruthenium, or the like.

The term "linking compound" or "linking group" as used herein generally refers to a hydrocarbonaceous moiety. A linking compound is capable of reacting, and a linking group may have ...

LEVEL 1 - 3 OF 68 PATENTS

5,601,934

<=2> GET 1st DRAWING SHEET OF 1

Feb. 11, 1997

Memory disk sheet stock and method

INVENTOR: Bartges, Charles W., Delmont, Pennsylvania Baumann, Stephen E., Penn Hills, Pennsylvania Hyland, Jr., Robert W., Oakmont, Pennsylvania Jensen, Craig L., Pittsburgh, Pennsylvania Tarcy, Gary P., Plum, Pennsylvania Vinnedge, K. Dean, Bettendorf, Iowa Skeen, Troy C., Bettendorf, Iowa

#### DETDESC:

... automatically grouped with this same series of elements even though it often performs the same function as scandium, or other "true" rare earths in an alloy composition. It is believed that minor amounts of still other rare earths, like erbium, thulium, lutetium, ytterbium, or another rare earth "act-alike", like hafnium, may be substituted for, or possibly even combined with scandium (or with each other) in varying quantities to achieve the ...

LEVEL 1 - 4 OF 68 PATENTS

5,593,951

<=2> GET 1st DRAWING SHEET OF 4

Jan. 14, 1997

Epitaxy of high T[C] superconductors on silicon

INVENTOR: Himpsel, Franz J., Mt. Kisco, New York

... as well as to understand the basic mechanisms for superconductivity in this class of materials.

Bednorz and Mueller first showed superconducting behavior in mixed copper-oxides, typically including rare earth and/or rare earth-like elements and alkaline earth elements, for example La, Ba, Sr, . . . , and having a perovskite-like structure.

Materials including the so called "1-2-3" phase in the Y-Ba-Cu-O ...

DETDESC:

... EMBODIMENTS

A technique is provided for depositing high T[c] superconducting copper-oxide based materials epitaxially on Si (001). Typically, these classes of superconducting materials include a rare earth or rare earth-like element and/or an alkaline earth element. Representative formulas for such materials are the following:

(A[1- chi]B chi)2Cu omicron [4- epsilon]

and

A1B2Cu30[7- epsilon ]

where A is a trivalent element (e.g., ...

... in the art that the present invention applies to epitaxial structures including silicon (001) surfaces and any copper oxide superconductor thereon. Thus, the teaching of this invention can include copper-oxide based compositions having any combinations of rare earth or rare earth-like elements and/or alkaline earth elements as well as copper oxide superconductors which do not contain rare earth elements. Further, it will be apparent to those of skill in the art that the Si (001) surface is ...

LEVEL 1 - 5 OF 68 PATENTS

5,573,574

Nov. 12, 1996

Electrorefined aluminium with a low content of uranium, thorium and rare earths

INVENTOR: Leroy, Michel, St. Egreve, France

SUM:

... applications specifies a minimum A1 content of above 99.9995%, (and even sometimes above 99.9997%) and a U + Th content of less than 1 ppb, and even sometimes less than 0.3 or 0.1 ppb.

Rare earths, some of which, like samarium, have a significant alpha radioactivity, are also undesirable. By way of example, 10 ppb of natural samarium emits as many alpha particles as 0.1 ppb of uranium 238. The high purity ...

LEVEL 1 - 6 OF 68 PATENTS

5,569,759

<=2> GET 1st DRAWING SHEET OF 25

Oct. 29, 1996

Water soluble texaphyrin metal complex preparation

INVENTOR: Sessler, Jonathan L., Austin, Texas Hemmi, Gregory W., Austin, Texas Mody, Tarak D., Austin, Texas

**DETDESC:** 

... C), 10.24 (s, 2 H, ArH), 12.23 (s, 2 H, CH=N); UV/vis: lambda [max 1420.0, 477.5, 730.0; FAB MS M< + > 811.

Other lanthanide and rare earth-like metal complexes may be synthesized including the Gd< + 3>, Lu< + 3>, La< + 3>, In< + 3>, and Dy< + 3>complexes.

EXAMPLE 4

Synthesis of B2T2 TXP, see FIGS. 7A and ... LEVEL 1 - 7 OF 68 PATENTS

5,567,564

<=2> GET 1st DRAWING SHEET OF 7

Oct. 22, 1996

Liquid development composition having a colorant comprising a stable dispersion of magnetic particles in an aqueous medium

INVENTOR: Ziolo, Ronald F., Webster, New York

#### DETDESC:

... described in U.S. Pat. No. 4,474,886 to Ziolo. Examples of the precursor ions which may be used includes those derivable from transition metal ions, such as iron, cobalt, nickel, manganese, vanadium, chromium, rare earths and the like. In the case of a non-magnetic colloid, this may include ions of, for example, sulfur, selenium, gold, barium, cadmium, copper, silver, manganese, molybdenum, zirconium, gallium, arsenic, indium, tin, ...

... ions which can be incorporated into the resin beads to form both single-domain and multidomain magnetic particles include: those derivable from transition metal ions, such as iron, cobalt, nickel, manganese, vanadium, chromium, rare earths and the like. These ions generally exist in the form of chlorides of the metal involved such as ferrous chloride, ferric chloride, copper chloride, nickel chloride, and the like. The corresponding iodides, bromides and fluorides may also be suitable. LEVEL 1 - 8 OF 68 PATENTS

5,554,428

Sep. 10, 1996

Memory disk sheet stock and method

INVENTOR: Bartges, Charles W., Delmont, Pennsylvania Hayland, Jr., Robert W., Oakmont, Pennsylvania Jensen, Craig J., Pittsburgh, Pennsylvania Baumann, Steven F., Penn Hills, Pennsylvania (Rule 47 Application)

... automatically grouped with this same series of elements even though it often performs the same function as scandium, or other "true" rare earths in an alloy composition. It is believed that minor amounts of still other rare earths, like erbium, thulium, lutetium, ytterbium, or another rare earth "act-alike", like hafnium, may be substituted for, or possibly even combined with scandium (or with each other) in varying quantities to achieve the ... LEVEL 1 - 9 OF 68 PATENTS

5,504,205

<=2> GET 1st DRAWING SHEET OF 25 

INVENTOR: Sessler, Jonathan L., Austin, Texas Hemmi, Gregory W., Austin, Texas Mody, Tarak D., Austin, Texas

# DETDESC:

... 2H, CH=C), 10.24 (s, 2H, ArH), 12.23 (s, 2H, CH=N); UV/vis: lambda max 420.0, 477.5, 730.0; FAB MS M< + > 811.

Other lanthanide and rare earth-like metal complexes may be synthesized including the Gd< + 3> , Lu< + 3> , La< + 3> , In< + 3 > and Dy< + 3 > complexes. EXAMPLE 4

Synthesis of B2T2 TXP, see FIG. 7.

**PAGE** 

LEVEL 1 - 10 OF 68 PATENTS

5,491,224

Feb. 13, 1996

Direct label transaminated DNA probe compositions for chromosome identification and methods for their manufacture

INVENTOR: Bittner, Michael L., 1768 Brookdale Rd., Naperville, Illinois 60563 Morrison, Larry E., 21 W. 559 Kensington Rd., Glen Ellyn, Illinois 60137 Legator, Mona S., 6540 N. Francisco, Chicago, Illinois 60645

#### DETDESC:

... capable of reacting, and a fluorophore group may have already reacted, with a linking group. A fluorescent compound may include an organic chelator which binds a luminescent inorganic ion such as a rare earth like terbium, europium, ruthenium, or the like.

The term "linking compound" or "linking group" as used herein generally refers to a hydrocarbonaceous moiety. A linking compound is capable of reacting, and a linking group may have ...

LEVEL 1 - 11 OF 68 PATENTS

5,475,104

<=2> GET 1st DRAWING SHEET OF 26

Dec. 12, 1995

Water soluble texaphyrin metal complexes for enhancing relaxivity

INVENTOR: Sessler, Jonathan L., Austin, Texas Hemmi, Gregory W., Austin, Texas Mody, Tarak D., Austin, Texas

#### DETDESC:

... 2H, CH=C), 10.24 (s, 2H, ArH), 12.23 (s, 2H, CH=N); UV/vis lambda max 420.0, 477.5, 730.0; FAB MS M< + > 811.

Other lanthanide and rare earth-like metal complexes may be synthesized including the Gd< + 3> , Lu< + 3> , La< + 3> , In< + 3 > and Dy< + 3 > complexes. Married and the bridge of the state of the s

EXAMPLE 4

Synthesis of B2T2 TXP, see FIGS. 7A and ...

LEVEL 1 - 122OF 68 PATENTS

<=2> GET 1st DRAWING SHEET OF 51

Oct. 10, 1995

# Hydroxylated texaphyrins

INVENTOR: Sessler, Jonathan L., Austin, Texas Mody, Tarak D., Sunnyvale, California Hemmi, Gregory W., Sunnyvale, California Kral, Vladimir, Na Kozaaoa, Czechoslovakia

#### DETDESC:

... 2H, CH=C), 10.24 (s, 2H, ArH), 12.23 (s, 2H, CH=N); UV/vis: lambda max 420.0, 477.5, 730.0; FAB MS M< + > 811.

Other lanthanide and rare earth-like metal complexes may be synthesized in a similar manner including the La< + 3> , Nd< + 3> , Sm< + 3> , Eu< + 3> , Gd< + 3> , Dy< + 3 > and Tm< + 3 > complexes. PAGE

LEVEL 1 - 13 OF 68 PATENTS

5,451,576

<=2> GET 1st DRAWING SHEET OF 26

Sep. 19, 1995

Tumor imaging and treatment by water soluble texaphyrin metal complexes

INVENTOR: Sessler, Jonathan L., Austin, Texas Hemmi, Gregory W., Austin, Texas Mody, Tarak D., Austin, Texas

## **DETDESC:**

... 2H, CH=C), 10.24 (s, 2H, ArH), 12.23 (s, 2H, CH=N); UV/vis: lambda max 420.0, 477.5, 730.0; FAB MS M< + > 811.

Other lanthanide and rare earth-like metal complexes may be synthesized including the Gd< + 3> , Lu< + 3> , La< + 3> , In< + 3> and Dy< + 3 > complexes. EXAMPLE 4

Synthesis of B2T2 TXP, see FIGS. 7A and ... LEVEL 1 - 14 OF 68 PATENTS

5,447,906

Sep. 5, 1995

Thin film high TC oxide superconductors and vapor deposition methods for making the same

INVENTOR: Chaudhari, Praveen, Briarcliff Manor, New York Gambino, Richard J., Yorktown Heights, New York Koch, Roger H., Amawalk, New York Lacey, James A., Mahopac, New York Laibowitz, Robert B., Peekskill, New York Viggiano, Joseph M., Wappingers Falls, New York

# ABST:

... films are produced by vapor depositions processes using pure metal sources for the metals in the superconducting compositions, where the metals include multi-valent nonmagnetic transition metals, rare earth elements and/or rare earth-like elements and alkaline earth-elements. The substrate is exposed to oxygen during vapor deposition, and, after formation of the film, there is at least one annealing step in an oxygen ambient and slow cooling over several

hours to room to perature. The substrates chosen we not critical as long as they are not adversely reactive with the superconducting oxide film. Transition metals include Cu, Ni, Ti and V, while the rare earth-like elements include Y, Sc and La. The alkaline earth elements include Ca, Ba and Sr.

... material in the last decade, wherein the critical transition temperature T c at which the material becomes superconducting was increased substantially.

Bednorz and Mueller described copper oxide material including a rare earth element, or rare earth-like element, where the rare earth element could be substituted for by an alkaline earth element such as Ca, Ba or Sr.

The work of Bednorz and Mueller has led to intensive investigation in many laboratories in ...

... 40o K. and methods for making these films, where the films exhibit perovskite-like structure.

It is another object of this invention to provide transition metal oxide superconductive films including a rare earth element, or rare earth-like element, where the films exhibit superconductivity at temperatures greater than 400 K., and methods for making these films.

It is another object of the present invention to provide films having the nominal composition ABO 3 - y or ABO y ...

... provide superconductive oxide films having the nominal composition AB2 Cu30 9 - y , and methods for making these films, where the films are superconducting at temperatures in excess of 400 K. and A is a rare earth or rare earth-like element, B is an alkaline earth element, and y is sufficient to satisfy valence demands of the composition.

Pat. No. 5447906, \*

It is another object of the present invention to provide smooth, continuous copper oxide superconducting films having a perovskite-like ...

... films being smooth and continuous and exhibiting substantial compositional uniformity. In particular, the films are comprised of transition metal oxides containing a superconducting phase, and typically include a rare earth element or rare earth-like element. These rare earth-like elements include Y, Sc and La. Additionally, the rare earth or rare earth-like elements can be substituted for by an alkaline earth element selected from the group consisting of Ca, Ba, and Sr. The transition metals are multi-valent, non-magnetic elements selected from the group consisting of ...

... especially a T c in excess of liquid nitrogen temperatures. These films are characterized by the presence of a transition metal oxide and typically by the presence of a rare earth element and/or a rare earth-like element which can be substituted for by an alkaline earth. The transition metal element is a multi-relent nonmagnetic element while the alkaline earth element is selected from the group consisting of Ca, Ba, and Sr. The rare earth-like elements include Y, Sc, and La. The nonmagnetic transition metal is selected from the group consisting of Cu, Ni, Ti, and V. Of these, Cu is the most favorable, yielding film properties which are unique and unexpected.

In the further ...

LEVEL 1 - 15 OF 68 PATENTS

5,439,570

<=2>GET 1st DRAWING SHEET OF 26
Aug. 8, 1995

Water soluble texaphyrin metal complexes for singlet oxygen

INVENTOR: Sessler, Jonathan L., Austin, Texas Hemmi, Gregory W., Austin, Texas Mody, Tarak D., Austin, Texas

#### **DETDESC:**

... 2H, CH=C), 10.24 (s, 2H, ArH), 12.23 (s, 2H, CH=N); UV/vis: lambda max 420.0, 477.5, 730.0; FAB MS M< + > 811.

Other lanthanide and rare earth-like metal complexes may be synthesized including the Gd< + 3> , Lu< + 3> , La< + 3> , In< + 3 > and Dy< + 3 > complexes. EXAMPLE 4

Synthesis of B2T2 TXP, see FIGS. 7A and ... LEVEL 1 - 16 OF 68 PATENTS

5,432,171

<=2> GET 1st DRAWING SHEET OF 26

Jul. 11, 1995

Water soluble texaphyrin metal complexes for viral deactivation

INVENTOR: Sessler, Jonathan L., Austin, Texas Hemmi, Gregory W., Austin, Texas Mody, Tarak D., Austin, Texas

#### DETDESC:

... 2H, CH=C), 10.24 (s, 2H, ArH), 12.23 (s, 2H, CH=N); UV/vis: lambda max 420.0, 477.5, 730.0; FAB MS M< + > 811.

Other lanthanide and rare earth-like metal complexes may be synthesized including the Gd< + 3> , Lu< + 3> , La< + 3> , In< + 3 > and Dy< + 3 > complexes. EXAMPLE 4

Synthesis of B2T2 TXP, see FIGS. 7A and ... LEVEL 1 - 17 OF 68 PATENTS

5,362,582

Nov. 8, 1994

## Battery separator

INVENTOR: Chang, Victor S., Ellicott City, Maryland Hartwig, Richard C., Laurel, Maryland Lundquist, Joseph T., Gilroy, California Parham, Marc E., Bedford, Massachusetts Kung, James K., Lexington, Massachusetts Avtges, James A., Belmont, Massachusetts Laccetti, Anthony J., North Andover, Massachusetts

#### MII2

... say the particulate filler must be inert with respect to such end use battery environment. Therefore, alkali insoluble particulate such as zirconia and titanium dioxide (preferred), oxides, hydroxides and carbonates of calcium, magnesium, iron, rare earth and the like should be used only in sheet products which ultimately are formed into battery separators for alkaline batteries. Similarly, acid insoluble particulates such as silica (a precipitated silica is preferred), and the like should be ...

LEVEL 1 - 18 OF 68 PATENTS

5,358,659

#### <=2> GET 1st DRAWING SHEET OF 5

Oct. 25, 1994

Magnetic materials with single-domain and multidomain crystallites and a method of preparation

INVENTOR: Ziolo, Ronald F., Webster, New York

#### DETDESC:

... Ions which can be incorporated into the resin beads to form both single-domain and multidomain magnetic particles include: those derivable from transition metal ions, such as iron, cobalt, nickel, manganese, vanadium, chromium, rare earths and the like. These ions generally exist in the form of chlorides of the metal involved such as ferrous chloride, ferric chloride, copper chloride, nickel chloride, and the like. The corresponding iodides, bromides and fluorides may also be suitable. ...

> PAGE 20

LEVEL 1 - 19 OF 68 PATENTS

5,322,756

<=2> GET 1st DRAWING SHEET OF 3

Jun. 21, 1994

Magnetic fluids and method of preparation

INVENTOR: Ziolo, Ronald F., Webster, New York

#### **DETDESC:**

... several different ions including ferrous or ferric ions. Examples of the precursor ions which may be used includes those derivable from transition metal ions, such as iron, cobalt, nickel, manganese, vanadium, chromium, rare earths and the like. These ions generally exist in the form of chlorides of the metal involved, such as ferrous chloride, ferric chloride, copper chloride, nickel chloride, and the like. The corresponding iodides, bromides and fluorides may also be suitable. ...

LEVEL 1 - 20 OF 68 PATENTS

5,304,966

<=2> GET 1st DRAWING SHEET OF 4

Apr. 19, 1994

Method of adjusting a frequency response in a three-conductor type filter device

INVENTOR: Hino, Seigo, Nagoya, Japan Ito, Kenji, Nagoya, Japan

... each other. Each of the dielectric substrates 1 and 2 may be of dielectric ceramic material having a high dielectric constant and a lower dielectric loss such as BaO-TiO2, BaO-TiO2-rare earth or the like. The lower dielectric substrate 1 is provided with an external ground conducting layer 3 on the peripheral portion and bottom surface thereof. Similarly, the upper dielectric substrate 2 is provided with an external ground conducting layer 4 on the ...

# DETDESC:

... assembling of the filter. Each of the dielectric substrates 21 and 22 may be of dielectric ceramic material having a high dielectric constant and a lower

dielectric loss such as BaO-TiO2, BaO-TiO2-rare earth or the like. The lower dielectric substrate 21 is provided with an external ground conductor layer 23 on the peripheral portion and outer surface thereof. Similarly, the upper dielectric substrate 22 is provided with an external ground conductor layer 24 on the ...

LEVEL 1 - 21 OF 68 PATENTS

5,296,458

<=2> GET 1st DRAWING SHEET OF 4

Mar. 22, 1994

Epitaxy of high T c superconducting films on (001) silicon surface

INVENTOR: Himpsel, Franz J., Mt. Kisco, New York

SUM:

... as well as to understand the basic mechanisms for superconductivity in this class of materials.

Bednorz and Mueller first showed superconducting behavior in mixed copper-oxides, typically including rare earth and/or rare earth-like elements and alkaline earth elements, for example La, Ba, Sr, . . . , and having a perovskite-like structure. Materials including the so called "1-2-3" phase in the Y-Ba-Cu-O . . .

#### DETDESC:

# ... EMBODIMENTS

A technique is provided for depositing high T c superconducting copper-oxide based materials epitaxially on Si (001). Typically, these classes of superconducting materials include a rare earth or rare earth-like element and/or an alkaline earth element. Representative formulas for such materials are the following:

(A 1 - x B x) 2CuO4 - epsilon and A1B2Cu3O 7 - epsilon

where A is a trivalent element (e.g., La, Y, and ...

... in the art that the present invention applies to epitaxial structures including silicon (001) surfaces and any copper oxide superconductor thereon. Thus, the teaching of this invention can include copper-oxide based compositions having any combinations of rare earth or rare earth-like elements and/or alkaline earth elements as well as copper oxide superconductors which do not contain rare earth elements. Further, it will be apparent to those of skill in the art that the Si (001) surface is ...

LEVEL 1 - 22 OF 68 PATENTS

\_\_ -

5,291,162

<=2> GET 1st DRAWING SHEET OF 7

Mar. 1, 1994

Method of adjusting frequency response in a microwave strip-line filter device

INVENTOR: Ito, Kenji, Nagoya, Japan Shimizu, Hiroyuki, Nagoya, Japan Oguchi, Hotaka, Nagoya, Japan

SUM:

... type which comprises a pair of dielectric cubstrates le and 1b made of dielectric ceramic material having a high dielectric constant and a lower dielectric loss such as BaO-TiO2 or BaO-TiO2-rare earth or the like, the

· 金.

dielectric substrates is and 1b being stacked to each other. The dielectric substrates 1a and 1b are provided with external ground conducting layers 2a and 2b on the peripheral portion and bottom surface thereof, respectively. On the upper ...

# **DETDESC:**

... assembling of the filter. Each of the dielectric substrates 11 and 12 may be of dielectric ceramic material having a high dielectric constant and a lower dielectric loss such as BaO-TiO2, BaO-TiO2-rare earth or the like. The lower dielectric substrate 11 is provided with an external ground conducting layer 13 on the peripheral portion and outer surface thereof. Similarly, the upper dielectric substrate 12 is provided with an external ground conducting layer 14 on the ...

... a pair of piezoelectric substrates 11 and 12 each of which may be of dielectric ceramic material having a high dielectric constant and a lower dielectric loss such as BaO-TiO2, BaO-TiO2-rare earth or the like. The dielectric substrates 11 and 12 are provided with external ground conducting layers 13 and 14 on the peripheral portions and outer surfaces thereof, respectively. These ground conducting layers 13 and 14 may be formed by ...

LEVEL 1 - 23 OF 68 PATENTS

5,278,140

<=2> GET 1st DRAWING SHEET OF 5

Jan. 11, 1994

Method for forming grain boundary junction devices using high T c superconductors

INVENTOR: Chaudhari, Praveen, Briarcliff Manor, New York
Chi, Cheng-Chung J., Yorktown Heights, New York
Dimos, Duane B., Montclair, New Jersey
Mannhart, Jochen D., Metzingen, New York, Federal Republic of Germany
Tsuei, Chang C., Chappaqua, New York

#### SUM:

... as well as to understand the basic mechanisms for superconductivity in this class of materials.

Bednorz and Mueller first showed superconducting behavior in mixed copper-oxides, typically including rare earth and/or rare earth-like elements and alkaline earth elements, for example La, Ba, Sr, . . . , and having a perovskite-like structure. Materials including the so called "1-2-3" phase in the Y-Ba-Cu-O . . .

... excess of about 30o K are generally known as "high T c superconductors", and will be referred to in that manner throughout the specification. This designation is meant to include both the materials having rare earth or rare earth-like elements in their crystalline structure, as well as the more recently reported materials which do not contain rare earth or rare earth-like elements. Generally, all these materials are copper oxide based superconductors having Cu-O planes that appear to be primarily responsible for carrying the supercurrents, where the copper oxide planes are separate or in groups separated by the ...

LEVEL 1 - 24 OF 68 PATENTS

5,252,720

<=2> GET 1st DRAWING SHEET OF 25

Oct. 12, 1993

Metal complexes of water soluble temphyrins-

INVENTOR: Sessler, Jonathan L., Austin, Texas

Hemmi, Gregory W., Austin, Texas Mody, Tarak D., Austin, Texas

#### **DETDESC:**

... 2H, CH=C), 10.24 (s, 2H, ArH), 12.23 (s, 2H, CH=N); UV/vis: lambda max 420.0, 477.5, 730.0; FAB MS M< + > 811.

Other lanthanide and rare earth-like metal complexes may be synthesized including the Gd< + 3> , Lu< + 3> , La< + 3> , In< + 3 > and Dy< + 3 > complexes. EXAMPLE 4

Synthesis of B2T2 TXP, see FIG. 7

**PAGE** 

LEVEL 1 - 25 OF 68 PATENTS

5,235,298

<=2> GET 1st DRAWING SHEET OF 2

Aug. 10, 1993

Temperature compensated stripline filter for microwaves

INVENTOR: Banno, Hisao, Nagoya, Japan Nishiki, Masahiro, Nagoya, Japan

SUM:

... 4,785,271 and Japanese Patent Prepublication No. 62-263702.

With the microwave stripline filter of the abovementioned type, generally, each dielectric ceramic substrate is made of ceramic material such as BaO-TiO2, BaO-TiO2-rare earth or the like.

However, there is disadvantage that the commonly used ceramic material has a resonant frequency which is decreased as the temperature is risen because the temperature coefficient of the resonant frequency is of a negative characteristic.

It is therefore an object of the present invention to provide a stripline ... LEVEL 1 - 26 OF 68 PATENTS

5,188,809

<=2> GET 1st DRAWING SHEET OF 4

Feb. 23, 1993

Method for separating coke from a feed mixture containing zirconium and radioactive materials by flotation process

INVENTOR: Crocker, William A., Salem, Oregon Haygarth, John C., Corvallis, Oregon Riesen, Jon A., Albany, Oregon Peterson, John R., Salem, Oregon

DETDESC:

... radium removal.

# <=2> GET 1st DRAWING SHEET OF 5

Nov. 10, 1992

Grain boundary junction devices using high T c superconductors

INVENTOR: Chaudhari, Praveen, Briarcliff Manor, New York
Chi, Cheng-Chung J., Yorktown Heights, New York
Dimos, Duane B., Upper Montclair, New Jersey
Mannhart, Jochen D., Metzingen, New York, Federal Republic of Germany
Tsuei, Chang C., Chappaqua, New York

#### SUM:

... as well as to understand the basic mechanisms for superconductivity in this class of materials.

Bednorz and Mueller first showed superconducting behavior in mixed copper-oxides, typically including rare earth and/or rare earth-like elements and alkaline earth elements, for example La, Ba, Sr, . . . , and having a perovskite-like structure. Materials including the so called "1-2-3" phase in the Y-Ba-Cu-O . . .

- ... excess of about 30o K. are generally known as "high T c superconductors", and will be referred to in that manner throughout the specification. This designation is meant to include both the materials having rare earth or rare earth-like elements in their crystalline structure, as well as the more recently reported materials which do not contain rare earth or rare earth-like elements. Generally, all these materials are copper oxide based superconductors having Cu-O planes that appear to be primarily responsible for carrying the supercurrents, where the copper oxide planes are separate or in groups separated by the ...
- ... [\*4] copper oxide material having a superconducting onset temperature greater than 77 K.
- [\*5] 5. The device of claim 4, where said superconducting material includes an atom selected from the group consisting of rare earth atoms and rare earth-like atoms.
- [\*6] 6. The device of claim 4, where said superconducting material includes an alkaline earth atoms.
- [\*7] 7. The device of claim 4, where said superconducting material includes bismuth.
  - [\*8] 8. The device of claim 1, where ... LEVEL 1 28 OF 68 PATENTS

5,160,482

<=2> GET 1st DRAWING SHEET OF 8

Nov. 3, 1992

Zirconium-hafnium separation and purification process

INVENTOR: Ash, Kenneth C., Corvallis, Oregon
Crocker, William A., Salem, Oregon
Haygarth, John C., Corvallis, Oregon
Lee, David R., Lebanon, Oregon
Morris, Donald, Corvallis, Oregon
Peterson, John R., Salem, Oregon
Riesen, Jon A., Albany, Oregon
Yih, Robert S., Salem, Oregon

#### DETDESC:

... system or solution.

b) Sodium sulfate or any other source of soluble sulfate is then added in excess of the concentration of the barium plus radium ion equivalents and any other cations which might combine with the sulfate ions, i.e. calcium, rare earths, or the like. If the solution is cold, it should be heated and a digestion allowed to take place which can range from a fairly short time up to hours or days. The preferred digestion period would be a few hours with ...

LEVEL 1 - 29 OF 68 PATENTS

5,112,795

May 12, 1992

Supported silver catalyst, and processes for making and using same

INVENTOR: Minahan, David M., Cross Lanes, West Virginia Thorsteinson, Erlind M., Charleston, West Virginia Liu, Albert C., Charleston, West Virginia

#### SUM:

... metal promoter employed is not critical and may include the one or more alkali metals; one or more alkaline earth metals; or one or more other promoters, such as thallium, gold, tin, antimony, rare earths and the like. The catalysts produced are said to be equally as efficient as catalysts produced by coincidental methods of preparation.

Supported, silver-containing, alkylene oxide catalysts often include one or more metal- ...

LEVEL 1 - 30 OF 68 PATENTS

5,084,684

<=2> GET 1st DRAWING SHEET OF 5

Jan. 28, 1992

Method of adjusting a frequency response in a three-conductor type filter device

INVENTOR: Shimizu, Hiroyuki, Nagoya, Japan Ito, Kenji, Nagoya, Japan Wakita, Naomasa, Nagoya, Japan

#### SUM:

... each other. Each of the dielectric substrates 1 and 2 may be of dielectric ceramic material having a high dielectric constant and a lower dielectric loss such as BaO-TiO2, BaO-TiO2-rare earth or the like. The lower dielectric substrate 1 is provided with an external ground conducting layer 3 on the peripheral portion and bottom surface thereof. Similarly, the upper dielectric substrate 2 is provided with an external ground conducting layer 4 on the ...

#### **DETDESC:**

... assembling of the filter. Each of the dielectric substrates 11 and 12 may be of dielectric ceramic material having a high dielectric constant and a lower dielectric loss such as BaO-TiO2, BaO-TiO2-rare earth or the like. The lower dielectric substrate 11 is provided with a ground conducting layer 13 on the lower or outer surface thereof. Similarly, the upper dielectric substrate 12 is provided with a ground conducting layer 14 on the upper or ...

LEVEL 1 - 31 OF 68 PATENTS

Jan. 28, 1992

Molten metal containment vessel with rare earth oxysulfide protective coating thereon and method of making same

INVENTOR: Krikorian, Oscar H., Danville, California Curtis, Paul G., Tracy, California

#### SUM:

... same. More particularly, this invention relates to an improved containment vessel for molten metals formed by coating at least the inside surface of a containment vessel with an oxysulfide or sulfide of a rare earth or rare earth-like element.

Molten metals such as uranium, plutonium, aluminum, and calcium are usually contained in vessels or crucibles made from graphite or a refractory metal such as, for example, niobium, tantalum, molybdenum, or tungsten. ...

... in which wetting of the vessel's surfaces by molten metal is inhibited by coating the surfaces of at least the inner walls of the containment vessel with one or more compounds comprising an oxysulfide of a rare earth or a rare earth-like element to inhibit such wetting and or adherence by the molten metal.

It is a further object of this invention to provide a method for making an improved molten metal containment vessel in which wetting of the surfaces by ...

#### DETDESC:

... rare earth oxysulfide or sulfide compound include the lanthanide elements La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu; as well as the rare earth-like elements Sc and Y; and actinides such as Th and U. The term "rare earth" and "rare earth elements", as used herein, are therefore intended to define any of the above listed elements.

The rare earth oxysulfide and sulfide coatings of the ... LEVEL 1 - 32 OF 68 PATENTS

5,075,653

Dec. 24, 1991

Method of adjusting a frequency response in a three-conductor type filter device

INVENTOR: Ito, Kenji, Nagoya, Japan Shimizu, Hiroyuki, Nagoya, Japan

#### SUM:

... each other. Each of the dielectric substrates 1 and 2 may be of dielectric ceramic material having a high dielectric constant and a lower dielectric loss such as BaO-TiO2, BaO-TiO2-rare earth or the like. The lower dielectric substrate 1 is provided with an external ground conducting layer 3 on the peripheral portion and bottom surface thereof. Similarly, the upper dielectric substrate 2 is provided with an external ground conducting layer 4 on the ...

#### **DETDESC:**

... assembling of the filter. Each of the dielectric substrates 11 and 12 may be of dielectric ceramic material having a high dielectric constant and a lower dielectric loss such as BaO-TiO2, BaO-TiO2-rare earth or the like. The lower dielectric substrate 11 is provided with an external ground conducting layer 13 on the peripheral portion and outer surface thereof. Similarly, the upper dielectric substrate 12 is provided with an external ground conducting layer 14 on the ...

5,066,934

## <=2> GET 1st DRAWING SHEET OF 6

Nov. 19, 1991

Method of adjusting a frequency response in a stripline filter device

INVENTOR: Ito, Kenji, Nagoya, Japan Shimizu, Hiroyuki, Nagoya, Japan Wakita, Naomasa, Nagoya, Japan

#### SUM:

... each other. Each of the dielectric substrates 1 and 2 may be of dielectric ceramic material having a high dielectric constant and a lower dielectric loss such as BaO-TiO2, BaO-TiO2-rare earth or the like. The lower dielectric substrate I is provided with an external ground conducting layer 3 on the peripheral portion and bottom surface thereof. Similarly, the upper dielectric substrate 2 is provided with an external ground conducting layer 4 on the ...

#### **DETDESC:**

... assembling of the filter. Each of the dielectric substrates 11 and 12 may be of dielectric ceramic material having a high dielectric constant and a lower dielectric loss such as BaO-TiO2, BaO-TiO2-rare earth or the like. The lower dielectric substrate 11 is provided with an external ground conducting layer 13 on the peripheral portion and outer surface thereof. Similarly, the upper dielectric substrate 12 is provided with an external ground conducting layer 14 on the ...

LEVEL 1 - 34 OF 68 PATENTS

5,045,289

<=2> GET 1st DRAWING SHEET OF 4

Sep. 3, 1991

Formation of rare earth carbonates using supercritical carbon dioxide

INVENTOR: Fernando, Quintus, Tucson, Arizona Yanagihara, Naohisa, Zacopan, New Mexico, Mexico Dyke, James T., Santa Fe, New Mexico Vemulapalli, Krishna, Tuscon, Arizona

# SUM:

... invention. This technique finds use in facilitating the extraction of these materials from rare earth containing mineral ores by providing a scheme for separating these particular rare earths from other rare earth and rare earth-like materials which do not react to form carbonates.

# 2. Description of the Prior Art

The rare earths, also known as the lanthanides or as lanthanons, and meaning here those elements having atomic numbers 57 to 71, are substances finding utility ...

## **DETDESC:**

... synthesis of rare earth carbonates from certain select rare earths in the trivalent (+3) state as normally found in, for example, rare earth oxides or hydroxides, from other rare earths or rare earths have materials. Rare earth-like materials are those compounds associated with, normally present in, or formed during the processing of, the various source ores from which the lanthanides are derived. These materials, while not true rare earths are analogous to the lanthanides in structure and behavior and are therefore of concern during processing and separation. Included among these rare earth-like

materials are compounds formed from the actinides, (elements of atomic numbers 89 to 103, such as thorium), titanium, yttrium, and zirconium. In general, these elements, which form the rare earth-like compounds, are present in their + 4 oxidation state; examples include ThO2 and ZrO2. The process of the invention has utility in the quantitative precipitation of the particular reactive lanthanides in the + 3 oxidation state and in the separation of these ...

... about 40o C. High yields of 95% or better are obtained in approximately one hour. These particular rare earth oxides or hydroxides can thus be readily separated from the oxides or hydroxides of rare earth or rare earth-like elements such as praseodymium (Pr), terbium (Tb), erbium (Er), ytterbium (Yb), zirconium (Zr), cerium (Ce), and thorium (Th) because these latter rare earth and rare earth-like oxides (or hydroxides) do not form carbonates under the above conditions despite the fact that some are in the trivalent state. It is believed that the oxides of these elements are particularly complex and as such do not readily react under the conditions of the invention.

This ...

Pat. No. 5045289, \*

... through appropriate valves and the reaction solution is then filtered. The solids which remain are then washed with deionized water and dried in air. These solids comprise both the rare earth materials which have reacted to form carbonates and also those rare earth and/or rare earth-like materials which did not react, or did not react significantly, and have thus remained in their oxide or hydroxide form.

The solid precipitate obtained above is next treated with a dilute acid such as HCl in a concentration of between 0.1 and 3.0M. Preferrably 0.5M HCl is used at ambient temperature and pressure. This acid treatment solubilizes the rare earth carbonates, leaving the unreacted rare earth and rare earth-like oxides and/or hydroxides behind in their solid form. The resultant solution is filtered and the carbonate fraction can be further broken down into individual rare earth carbonates by techniques such as ion exchange or ...

... La203(49.72%), Nd203(20.02%), Tb407(5.08%), Yb203(5.10%) and Th02(20.07%), a high degree of separation of La and Nd was obtained-namely, between 94.3% and 99.8%. Notably, the other rare earth or rare earth-like oxides in this mixture are among those which do not react to form carbonates with supercritical carbon dioxide or by the process of the invention.

The following example will illustrate and describe without limiting the invention. The example illustrates the carbonation process of the invention using essentially pure rare earth oxides. **EXAMPLE** 

Synthesis of Lanthanide Carbonates

The oxides of the following rare earths and rare earth-like materials, La203, CeO2, Pr6011, Nd203, Sm203, Eu203, Gd203, Tb407, Dy203, Ho203, Er203, Yb203 and ZrO2, were obtained from Alfa Division, Danvers, MA, and were 99.9% pure. The carbon dioxide used in this ...

LEVEL 1 - 35 OF 68 PATENTS

4,977,937

<=2> GET 1st DRAWING SHEET OF 4

Dec. 18, 1990

Multiple angle jointer and planer knives

INVENTOR: Hessenthaler, George D., 585 W .. 3900 South, #6, Murray, Utah 84123

DETDESC:

· -- -gibe or locking bars, not-shows, are tightened, the blade magnets 53 are selected to attract even minimally magnetic material, such as carbide. To provide such magnetic attraction the selected magnets should be very strong,

such as rare earth, or like magnets.

Like the jointer jig 40, a planer jig 60, shown in FIGS. 9 and 10 also utilizes magnets for maintaining blade positioning in a cylindrical cutterhead 61 ...

LEVEL 1 - 36 OF 68 PATENTS

4,962,086

<=2> GET 1st DRAWING SHEET OF 2

Oct. 9, 1990

High T c superconductor - gallate crystal structures

INVENTOR: Gallagher, William J., Ardsley, New York Giess, Edward A., Purdys, New York Gupta, Aranava, Valley Cottage, New York Laibowitz, Robert B., Peekskill, New York O'Sullivan, Eugene J., Peekskill, New York Sandstrom, Robert L., Chappaqua, New York

#### ABST:

High T c oxide superconductive films can be formed on gallate layers, where the gallate layers include a rare earth element or a rare earth-like element. Combinations of rare earth elements and rare earth-like elements can also be utilized. The superconductive films can be epitaxially deposited on these gallate layers to form single crystals or, in the minimum, highly oriented superconductive layers. Any high T c superconductive ...

#### SUM:

... materials having Cu-O planes therein which are responsible for carrying supercurrents in these materials. Epitaxial films of these high T c superconductors can be deposited on gallate substrates, where the substrates are rare earth gallates or rare earth-like gallates. These superconductor-substrate combinations are particularly suited for analog and digital signal processing devices including matched filters, correlators, Fourier transformers, spectrum analyzers, samplers, A/D converters, etc.

... high T c superconductors.

The high T c superconductors used with these gallate substrates are preferably those which include Cu-O and Cu-O like current carrying planes and can include rare earth and rare earth-like elements, as well as combinations of these elements. Also included are the non-rare earth high T c superconductors such as those having Bi-Sr-Ca-Cu-O compositions and Tl-Ba-Ca-Cu-...

... less than that when copper containing oxide superconductors are used. Lattice matching of the superconductor atomic spacing to the Ga-O plane is especially good with the copper oxide superconductors which form unique combinations with these gallates.

These rare earth and rare earth-like gallate substrates can be prepared in high quality crystal form and provide excellent lattice matches to the Cu-O based superconducting perovskites. This is important in device applications since for ...

DRWDESC:

BRIEF DESCRIPTION OF THE DRAWINGS

Pat. No. 4962086, \*

FIG. 1 illustrates a high T c superconducting film epitaxially deposited on a

FIG. 2 illustrates a structure including a high T c superconducting strip

line surrounded by a gallate lattice-matched insulator, and further including high ...

#### DETDESC:

... 10 has been deposited on the crystal substrate 12. A cooling means, if needed, is not shown but is well known in the art.

Substrate 12 is a gallate substrate comprised of a rare earth or rare earth-like element, gallium, and oxygen. Examples include LaGaO3 and NdGaO3. A mixed gallate can also be used, such as one prepared from La-Y solid solutions. This technique is used to provide different lattice ...

... for use in the substrate include elements 58-71 of the periodic table, and in particular, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu. The rare earth-like elements suitable for use in the gallate substrates include Y, La, Bi and Sc. As noted, combinations of these rare earth and rare earth-like elements can also be used.

For the copper oxide superconductors the rare earth elements Tb, Dy, Ho, Er, Tm, Yb, and Lu may not provide atomic spacings that give lattice ...

... one which in preferred form is characterized by Cu-O planes that are primarily responsible for carrying the supercurrents in these materials. They generally have a perovskite-related structure and can include rare earth and/or rare earth-like elements. These materials often include alkaline earth elements, as for example Ca, Ba, Sr, Mg, . . . An example of a 920 K. superconductor is the well known YBa2Cu3O 7 - x , which is the so-called " . . .

... be difficult to stabilize the approximately 1100 K. superconducting phase of Bi-Sr-Ca-Cu-O superconductors. However, a favorable epitaxial substrate chosen from the class of gallates including a rare earth or rare earth-like element may aid in stabilizing this and other high T c phases. A cut along the [110] orthorhombic unit cell of GdGaO3 would expose a surface with a favorable lattice match which ...

... While the unit cell of this superconducting thin film is rotated 450 with respect to the unit cell of the perovskite substrate, such rotation will not be needed for epitaxial matches of different superconductors to the rare earth and rare earth-like gallate substrates. One of skill in the art would use an orientation of the substrate such that good epitaxy and lattice matching will occur with the chosen superconducting film. In this example, the a and b axes are in the plane of the ...

... approximates a (100) cubic perovskite surface. With this as a guide, the substrate boule material is cut to provide the desired orientation.

It has been noted that the gallate substrates including a rare earth element or a rare earth-like element exhibit good hardness and tolerance to high temperatures. However, it may be preferable to process the superconducting film at temperatures less than the rhombohedral-orthorhombic transition of the substrate in order to maintain the slight orthorhombicity of the substrate.

Pat. No. 4962086, \*

# ... Lett. 58, 2684 (1987).

In the practice of this invention, highly oriented films of high T c oxide superconductors have been deposited on gallate substrates. These substrates are those which include at least one rare earth element or rare earth-like element. The superconducting epitaxial films are highly oriented and can approximate single crystals.

In the further practice of this invention, these high T-c oxide superconducting film-gallate substrate combinations are particularly suitable

A 19

... apparent to those of skill in the art that variations can be made therein without departing from the spirit and scope of the present invention. For example, the gallate substrate materials may include combinations of rare earth elements and rare earth-like elements, and may also be doped to slightly vary lattice parameters. Further, the superconductive films deposited on these substrates, while preferably being copper oxide-based superconductors, can include rare earth elements, rare earth-like elements, and alkaline earth elements. Still further, combinations of these elements may be present and, also, rare earth elements need not be present in the superconducting film.

The best epitaxial matches occur when the ... LEVEL 1 - 37 OF 68 PATENTS

4,882,718

<=2> GET 1st DRAWING SHEET OF 3

Nov. 21, 1989

Single-head, direct overwrite magneto-optic system and method

INVENTOR: Kryder, Mark H., Pittsburgh, Pennsylvania Shieh, Han-Ping D., Pittsburgh, Pennsylvania

#### DETDESC:

... domain will realign and not grow. Ferrimagnetic alloys including light rare earths such as gadolinium usually provide good mobility but generally require an approximately equal proportion of a heavy rare earth like terbium to increase coercivity to an effective operating level.

A preferred formulation (in atomic %) tested in the laboratory is as follows:

Gd13 Tb13 Fe59 Co15 having a compensation temperature of 900 ... LEVEL 1 - 38 OF 68 PATENTS

4,882,067

<=2> GET 1st DRAWING SHEET OF 1

Nov. 21, 1989

Process for the chemical bonding of heavy metals from sludge in the silicate structure of clays and shales and the manufacture of building and construction materials therewith

INVENTOR: Johnson, Barrett, Sunnyvale, California Rubenstein, Charles B., Los Gatos, California

## **DETDESC:**

... containing heavy metals which are generally considered to be toxic to humans and animal life, including arsenic, cobalt, cadmium, chromium, lead, nickel, selenium, thallium, zinc, magnesium, copper, antimony, barium, molybdenum, rare earths and the like and incidental organic toxins. In general, the invented process comprises a batch or continuous operation for the processing of industrial waste and contaminated water. The process developed as described in this patent is not ...

LEVEL 1 - 39 OF 68 PATENTS

4,806,328

Feb. 21, 1989

Method of manufacturing monolithic glass members

INVENTOR: Van Lierop, Joseph G., Eindhoven, Netherlands Bogemann, Arnoldus B. M., Eindhoven, Netherlands

Felder, Willy J. B., Vijlen, Netherlands Huizing, Albert, Eindhoven, Netherlands

SUM:

... example, to adjust the refractive index of the glass member obtained after densification of the gel at a given value and/or to control other physical properties. Examples of such compounds are alkoxy compounds of aluminium, titanium, boron, germanium, rare earths and the like, of which the alkoxy groups each generally do not comprise more than 4 carbon atoms. Nitrates, carbonates, acetates and other compounds which decompose easily while forming oxides, may optionally also be used. Fluorine ...

LEVEL 1 - 40 OF 68 PATENTS

4,775,820

<=2> GET 1st DRAWING SHEET OF 3

Oct. 4, 1988

Multilayer electroluminescent device

INVENTOR: Eguchi, Ken, Yokohama, Japan Kawada, Haruki, Kawasaki, Japan Nishimura, Yukuo, Sagamihara, Japan

SUM:

... composed of a material of EL function dispersed in a binder.

As the material of EL function, there have been known heretofore inorganic metal materials such as ZnS containing Mu, Cu, ReF3 (Re: rare earths) or the like as an activating agent, and the like.

In the case of a thin film type EL device, the structure is suitable for the following purposes, that is, a thin luminescent layer can be formed so as to ...

LEVEL 1 - 41 OF 68 PATENTS

4,734,338

<=2> GET 1st DRAWING SHEET OF 3

Mar. 29, 1988

Electroluminescent device

INVENTOR: Eguchi, Ken, Yokohama, Japan Kawada, Haruki, Kawasaki, Japan Nishimura, Yukuo, Sagamihara, Japan

SUM:

... layer composed of a material of EL function dispersed in a binder.

As the material of EL function, there have been heretofore inorganic metal materials such as ZnS containing Mn, Cu, ReF3 and (Re: rare earths) or the like as an activating agent, and the like.

In the case of a thin film type EL device, the structure is suitable for the following purposes, that is, a thin luminescent layer can be formed so as to ...

PAGE 46

LEVEL 1 - 42 OF 68 PATENTS

4,700,436

<=2> GET 1st DRAWING SHEET OF 4

Oct. 20, 1987

Magnetic fastener

INVENTOR: Morita, Tamao, 47-1, Arakawa 6-Chome, Arakawa-ku, Tokyo, Japan

SUM:

BACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates to the utilization of permanent magnets made of hard magnetic powder of ferrite, alnico, rare-earth and the like materials solidified with synthetic resin and then magnetized. More particularly, it relates to an improvement is magnetic material fastener means made of permanent magnet which is provided with magnetic plates at its magnetic poles.

2. Description of the Prior Art

PAGE

LEVEL 1 - 43 OF 68 PATENTS

4,681,625

<=2> GET 1st DRAWING SHEET OF 11

Jul. 21, 1987

Methods for simultaneously desulfurizing and degassing steels

INVENTOR: Wilson, William G., 820 Harden Dr., Pittsburgh, Pennsylvania 15229

SUM:

... difficult to get into solution and also those whose recoveries from their addition have been less than the amount added to the steel such as electrolytic manganese, ferro-niobium, ferro-tungsten and the like. The metals that may be added include aluminum, calcium, barium, rare earths and the like. The recovery of elements in the steel from additions of metals and ferro-alloys is reduced in many cases in conventional steel making technology by their contact with slags high in oxides such as iron ...

... [\*21] metals to be added in the tube to enhance desulfurization are those which are known to have the ability to reduce the oxygen content of the steel, but also have the ability to form sulfides which would float out of the steel into the slag which include magnesium, calcium, barium, rare earths and the like.

[\*22] 22. The method as claimed in claims 1 or 5 wherein the ferro-alloys and elemental metals to be added in the tube are those necessary to obtain the desired chemical analysis of the finished steel such as ferro- ...

LEVEL 1 - 44 OF 68 PATENTS

4,598,914

<=2> GET 1st DRAWING SHEET OF 10

Jul. 8, 1986

Sealing and bearing means by use of ferrofluid

INVENTOR: Furumura, Kyozaburo, Ninomiya, Japan Sugi, Hiromi, Fujisawa, Japan Murakami, Yasuo, Fujisawa, Japan Asai, Hiromitsu, Fujisawa, Japan

**DETDESC:** 

... polyamide resin, fluorine resin, polyethersulfone resin, polyphenylene

sulfide resin or the water. The magnetic material to be exixed with the aforesaid synthetic resin material is made of barium ferrite powder, strontium ferrite powder, rare earths or the like.

The mixture ratio of the synthetic resin and the aforesaid normal magnetic substance is different in case the magnet is used for bearing purposes and sealing purposes.

In case the magnet is employed as bearing, it is to have enough  $\dots$  LEVEL 1 - 45 OF 68 PATENTS

4,582,688

<=2> GET 1st DRAWING SHEET OF 1

Apr. 15, 1986

Process for recovery of mineral values

INVENTOR: Venkatesan, Valadi N., Arlington, Texas

#### **DETDESC:**

... present, molybdenum can be selectively leached from the ore utilizing a leaching solution containing sodium bicarbonate and oxygen. Thus, for example, substances such as vanadium, molybdenum, selenium, nickel, copper, uranium, the rare earths and the like may be recovered using the process of the present invention. The main criteria is that at least one of the minerals found in the ore may be solubilized without the solubilization of at least one other mineral.

Thus, the present ...

... part of the uranium is present as a refractory uranium-mineral complex. For example, other minerals found in the form of a uranium-mineral complex, include copper, nickel, thorium, scandium, the rare earths, and the like.

Uranium minerals frequently occur in the highly siliceous rocks and sedimentary deposits, generally as a mixture of the insoluble tetravalent form and the soluble hexavalent form. Uranium is also found in association with the silicates, ...

LEVEL 1 - 46 OF 68 PATENTS

4,570,692

<=2> GET 1st DRAWING SHEET OF 6

Feb. 18, 1986

Methods of pouring metal

INVENTOR: Wilson, William G., 820 Harden Dr., Pittsburgh, Pennsylvania 15229

#### **DETDESC:**

... teeming operation and good distribution throughout the entire ingot can be expected. When the stability of the oxides in the slags is high, even the most reactive alloys such as aluminum, titanium, zirconium, magnesum, calcium or rare earths and the like will be transferred to the steel from the slag with maximum retention of the alloying element in the metal being teemed. The addition of these alloys along with these stable oxides that will not react with these alloying elements, the elimination of the flow ...

LEVEL 1 - 47 OF 68 PATENTS

4,491,563

Jan. 1, 1985

Process for deodorizing a paraffinic hydrocarbon feedstock

INVENTOR: Reusser, Rout E., Bartlesville, Oklahoma Murtha, Timothy P., Bartlesville, Oklahoma Todd, Elizabeth A., Bartlesville, Oklahoma

#### **DETDESC:**

... examples are given to provide a better and more complete disclosure of this invention but should not be interpreted to limit its scope. EXAMPLE I

This example describes a typical catalyst preparation whereby NiO and a rare earth like CeO is deposited on a support. This general procedure is also described in U.S. Pat. No. 4,217,248 column 7, line 49 to column 8, line 41. Two hundred grams of  $13 \times \ldots$ 

LEVEL 1 - 48 OF 68 PATENTS

4,489,042

Dec. 18, 1984

Process for recovery of mineral values from subterranean formations

INVENTOR: Savins, Joseph G., Dallas, Texas Johnson, Warren F., Dallas, Texas

#### **DETDESC:**

... formations. However, it should be clear that the invention is applicable to the solution leaching of other mineral values capable of forming soluble reaction products with leaching solutions. Thus, for example, substances such as vanadium, molybdenum, nickel, copper, the rare earths and the like are recovered using the process of the present invention.

As an illustration, the leach chemistry of a uranium ore body can be described by the following equations using hydrogen peroxide (H2O2) as oxidant:

PAGE

LEVEL 1 - 49 OF 68 PATENTS

4,486,026

<=2> GET 1st DRAWING SHEET OF 10

Dec. 4, 1984

Sealing and bearing means by use of ferrofluid

INVENTOR: Furumura, Kyozaburo, Ninomiya, Japan Sugi, Hiromi, Fujisawa, Japan Murakami, Yasuo, Fujisawa, Japan Asai, Hiromitsu, Fujisawa, Japan

#### **DETDESC:**

43° 4117- ---

... polyamide resin, fluorine resin, polyethersulfone resin, polyphenylene sulfide resin or the like. The magnetic material to be mixed with the aforesaid synthetic resin material is made of barium ferrite powder, strontium ferrite powder, rare earths or the like.

The mixture ratio of the synthetic resin and the aforesaid normal magnetic substance is different in case the magnet is used for bearing purposes and sealing purposes.

In case the magnet is employed as bearing, it is to have enough ...

PAGE 54

LEVEL 1 - 50 OF 68 PATENTS

# <=2> GET 1st DRAWING SHEET OF 3

Nov. 6, 1984

Variable flux permanent magnets electromagnetic machine

INVENTOR: Parker, Rollin J., Greenville, Michigan

DETDESC:

... cylindrical housing 12 in which is mounted, by any appropriate convenient means, a cylindrical tubular stator 14 comprising high strength permanent magnets such as ceramic, or ceramic rare earth, cobalt-rare earth, or the like [magents] magnets. Each one of a pair of end cap members 16 and 18 fastened at an end of the housing 12 by bolts or screws 20 supports respectively an end magnet ring 22 an ...

LEVEL 1 - 51 OF 68 PATENTS

4,455,392

Jun. 19, 1984

Process for preparing a supported silver catalyst

INVENTOR: Warner, Glenn H., St. Albans, West Virginia Bhasin, Madan M., Charleston, West Virginia Lieberman, Bernard, Kew Gardens, New York

... as lithium, sodium, potassium, rubidium and/or cesium; one or more alkaline earth metals, such as, barium, magnesium and strontium; or one or more of the other known promoters, such as thallium, gold, tin, antimony and rare earths; and the like. For purposes of convenience, the catalyst preparation process of the invention is described below in terms of a silver-first method of preparation wherein the promoter is selected from among alkali metals, it being recognized that other promoters of ... LEVEL 1 - 52 OF 68 PATENTS

4,438,077

Mar. 20, 1984

Two stage selective oxidative leach method to separately recover uranium and refractory uranium-mineral complexes

INVENTOR: Tsui, Tien-Fung, Richardson, Texas

... least part of the uranium is present as a refractory uranium-mineral complex. For example, other minerals found in a uranium-mineral complex include copper, nickel, thorium, scandium, the rare earths, and the like.

Uranium minerals frequently occur in the highly siliceous rocks and sedimentary deposits, generally as a mixture of the insoluble tetravalent form and the soluble hexavalent form. Uranium is also found in association with the silicates, ...

LEVEL 1 - 53 OF 68 PATENTS

4,427,236

Jan. 24, 1984

In-situ uranium leaching

INVENTOR: Dotson, Billy J., Grand Prairie, Texas

· SHIP STITE - \*\*-

... be clear that the invention is applicable to the solution mining of other mineral values capable of forming soluble reaction products with carbonated leaching solutions. Thus, for example, substances such as vanadium, molybdenum, nickel, copper, the rare earths and the like are recovered using the process of the present invention.

Uranium minerals frequently occur in the highly siliceous rocks and sedimentary deposits, generally as a mixture of the insoluble quadrivalent form and the soluble sexivalent form. ...

LEVEL 1 - 54 OF 68 PATENTS

4,419,276

Dec. 6, 1983

Silver catalyst for the manufacture of ethylene oxide and a process for preparing the catalyst

INVENTOR: Bhasin, Madan M., Charleston, West Virginia Warner, Glenn H., St. Albans, West Virginia

SUM:

... as lithium, sodium, potassium, rubidium and/or cesium; one or more alkaline earth metals, such as, barium, magnesium and strontium; or one or more of the other known promoters, such as thallium, gold, tin, antimony and rare earths; and the like. For purposes of convenience, the catalyst preparation process of the invention is described below in terms of a silver-first method of preparation wherein the promoter is selected from among alkali metals, it being recognized that other promoters of ...

PAGE 59

LEVEL 1 - 55 OF 68 PATENTS

4,405,380

Sep. 20, 1983

High strength, low alloy steel with improved surface and mechanical properties

INVENTOR: Griffith, Cecil B., North Royalton, Ohio Thomas, Jerry D., North Olmsted, Ohio Demianczuk, Dionisyj W., Parma, Ohio Abraham, John K., Broadview Heights, Ohio Franklin, Joseph E., Medina, Ohio

#### **DETDESC:**

-J:\_

... present invention is directed to a steel with carbon in the range of 0.03 to 0.06%, the last being an upper limit which also appears crucial for attainment of so-called auto-sulfide-shape control and thus avoidance of the use of rare earths or the like with their consequent expense and tendency to produce unwanted non-metallic surface inclusions.

The base metal may thus consist of the defined composition, with manganese in the range of 0.2 to 0.6%, very preferably not more than 0.45%, while the ...

PAGE 60

LEVEL 1 - 56 OF 68 PATENTS

4,376,264

<=2> GET 1st DRAWING SHEET OF 6

Mar. 8, 1983

Method of checking the authenticity of papers and physically identifiable paper for use in said method

INVENTOR: Dokter, Hendrik D., Ugchelen, Netherlands Hildering, Roelof, Frederikslaan, Netherlands Mackor, Adrianus, Hollandsche Rading, Netherlands

SUM:

... be some which show a suitable ESR spectrum, although to the knowledge of the present inventors this has never been investigated. However, a further requirement is that a useful ESR spectrum should be obtained at room temperature. Many compounds of rare earths and the like show a useful ESR spectrum only at low temperatures, such as the temperature of liquid nitrogen, but of course an identification of banknotes and the like is hardly of any practical value, if it cannot be carried out at normal room ...

PAGE 61

LEVEL 1 - 57 OF 68 PATENTS

4,367,163

<=2> GET 1st DRAWING SHEET OF 1

Jan. 4, 1983

Silica-clay complexes

INVENTOR: Pinnavaia, Thomas J., East Lansing, Michigan Mortland, Max M., East Lansing, Michigan Endo, Tadashi, East Lansing, Michigan

#### DETDESC:

... be used as a catalyst support for various catalytically active metals such as a Group VIII metal such as platinum, palladium, nickel, iron or cobalt; molybdenum; tungsten; a rare-earth and the like. Moreover, the intercalated product can be used in admixture with other common adsorbents or matrix materials such as silica, alumina, silica-alumina hydrogel and the like. The catalysts which can be prepared by ...

LEVEL 1 - 58 OF 68 PATENTS

4,358,158

<=2> GET 1st DRAWING SHEET OF 1

Nov. 9, 1982

Solution mining process

INVENTOR: Showalter, William E., Seal Beach, California

## **DETDESC:**

... invention is applicable to the solution mining of other mineral values capable of forming soluble reaction products with the dilute carbonic acid leaching solution. Thus, for example, substances such as vandium, molybdenum, nickel, copper, the rare earths and the like can be recovered using the process of the present invention.

Uranium minerals frequently occur in the highly siliceous rocks and sedimentary deposits, generally as a mixture of the insoluble quadrivalent form and the soluble ...

LEVEL 1 - 59 OF 68 PATENTS

4,358,157

<=2> GET 1st DRAWING SHEET OF 1

Nov. 9, 1982

Solution mining process

INVENTOR: Showalter, William E., Seal Beach, California DETDESC: ... invention is applicable to the solution mining of other mineral values capable of forming soluble reaction products with the dilute carbonic acid leaching solution. Thus, for example, substances such as vanadium, molybdenum, nickel, copper, the rare earths and the like can be recovered using the process of the present invention. Uranium minerals frequently occur in the highly siliceous rocks and sedimentary deposits, generally as a mixture of the insoluble quadrivalent form and the soluble ... LEVEL 1 - 60 OF 68 PATENTS 4,328,079 <=2> GET 1st DRAWING SHEET OF 1 May 4, 1982 Method for pumping impurities, especially noble gases, from hydrogen or mixtures of hydrogen and its isotopes INVENTOR: Hemmerich, Johann, Stetternich, Federal Republic of Germany ... 2 is adjusted by the fluid within the chamber 13 to the temperature for the desired hydrogen partial pressure. In this variation, the cathodes are formed from hydride-forming metals and alloys, for example, rare earth and rare earth-like metals and binary and ternary alloys of them with the addition of transition metals like iron, nickel, cobalt, etc. Upon formation of the sputtered film 12, hydrogen and its isotopes form hydrides with the film by chemisorption that can ... LEVEL 1 - 61 OF 68 PATENTS 4,279,668 <=2> GET 1st DRAWING SHEET OF 7 Jul. 21, 1981 Directionally solidified ductile magnetic alloy INVENTOR: Kurz, Wilfried, Lausanne, California, Switzerland Glardon, Remi, Berkeley, California ... relates to a process for the fabrication of magnetic alloys for permanent magnets and to the magnetic bodies obtained by this process. More particularly the invention relates to ternary magnetic alloys consisting of rare-earth or rare-earth-like elements, cobalt and at least one metal selected from the group which consists of iron, nickel, aluminum, copper, molybdenum or manganese. Preferably the latter metal phase includes 0.1 to 10% (atomic) of the total alloy as ... LEVEL 1 - 62 OF 68 PATENTS 4,208,225 <=2> GET 1st DRAWING SHEET OF 6 Jun. 17, 1980 Directionally solidified ductile magnetic alloys magnetically hardened by precipitation hardening

INVENTOR: Kurz, Wilfried, Lausanne, Switzerland Glardon, Remi, Corseaux, Switzerland

SUM:

... relates to a process for the fabrication of magnetic alloys for permanent magnets and to the magnetic bodies obtained by this process.

More particularly the invention relates to ternary magnetic alloys consisting of rare-earth or rare earth-like elements, cobalt and at least one metal selected from the group which consists of iron, nickel, aluminum, copper, molybdenum or manganese.

BACKGROUND OF THE INVENTION

Ferromagnetic alloys of the cobalt/rare-earth type have a high energy ... LEVEL 1 - 63 OF 68 PATENTS

4,105,253

<=2> GET 1st DRAWING SHEET OF 1

Aug. 8, 1978

Process for recovery of mineral values from underground formations

INVENTOR: Showalter, William E., Seal Beach, California

DETDESC:

... be clear that the invention is applicable to the solution mining of other mineral values capable of forming soluble reaction products with carbonated leaching solutions. Thus, for example, substances such as vanadium, molybdenum, nickel, copper, the rare earths and the like are recovered using the process of the present invention.

Uranium minerals frequently occur in the highly siliceous rocks and sedimentary deposits, generally as a mixture of the insoluble quadrivalent form and the soluble sexivalent form. ...

LEVEL 1 - 64 OF 68 PATENTS

4,050,052

<=2> GET 1st DRAWING SHEET OF 1

Sep. 20, 1977

Electrical temperature measuring resistor structure, particularly for resistance thermometers

INVENTOR: Reichelt, Walter, Hanau, Germany, Federal Republic of Sauer, Gunter, Maintal, Germany, Federal Republic of

DETDESC:

... temperatures can be applied. This cover layer, shown in FIG. 2 schematically at 3, may consist for example of an epoxy resin, glass, or metal oxides of the group of aluminum, beryllium, thorium, rare earths, or the like. The cover layer 3 may be applied by vapor deposition, dusting, or spraying; its primary characteristics should be to be resistant against thermal and mechanical effects. The cover layer should additionally, preferably, provide ...

LEVEL 1 - 65 OF 68 PATENTS

4,014,706

Mar. 29, 1977

Solid solution ceramic materials

INVENTOR: Waldron, Robert D., Scottsdale, Arizona

SUM:

... dimensions of said structure and all physical and chemical properties of the solution are continuous functions of composition. The lattice symmetry may change within said composition range by uniform distortion of the structure as the composition changes.

Rare earth-like (metallic) elements as used herein means elements of atomic numbers 21, 39, and/or 57-71.

Yttrium earth (metallic) elements as used herein means elements of atomic numbers 39 and/or 64-71.

. . .

PAGE

LEVEL 1 - 66 OF 68 PATENTS

3,983,077

<=2> GET 1st DRAWING SHEET OF 2

Sep. 28, 1976

Process for making ceramic resistor materials

INVENTOR: Fuller, Peter G., Lakeville, Massachusetts Stoeckler, Hans A., Woonsocket, Rhode Island

#### DETDESC:

... invention also typically include additions of silicon oxide or manganese oxide or the like and other dopants typically incorporated in such ceramic compositions include lanthanum, cerium, dysprosium, and praesodymium as well as other rare earths and the like commonly used in ceramic resistor materials of positive temperature coefficient of resistivity. Typically, the ceramic titanate materials produced by the process are provided with stoichiometric or slightly titanium-rich compositions, the compositions preferably having an ...

LEVEL 1 - 67 OF 68 PATENTS

3,896,616

<=2> GET 1st DRAWING SHEET OF 1

Jul. 29, 1975

Process and apparatus

INVENTOR: Keith, Carl D., Summit, New Jersey Mooney, John J., Wyckoff, New Jersey

#### **DETDESC:**

... 0.1 to 1.5%. The catalytic element may contain, with or without the platinum group metals, one or more catalytic materials which may include, for example, chromium, manganese, vanadium, copper, iron, cobalt, nickel, rare earths, and the like.

The relative sizes of the initial and subsequent catalytic elements may be such that their volume ratio, i.e. the superficial volume of the subsequent catalyst to the initial catalyst, including void spaces within the catalytic masses, is often at least about ...

LEVEL 1 - 68 OF 68 PATENTS

3,791,143

<=2> GET 1st DRAWING SHEET OF 1

Feb. 12, 1974

PROCESS AND APPARATUS

INVENTOR: Keith, Carl D., Summit, New Jersey Mooney, John J., Wyckoff, New Jersey

#### **DETDESC:**

... 1.5 percent. The catalytic element may contain, with or without the platinum group metals, one or more catalytic materials which may include, for example, chromium, manganese, vanadium, copper, iron, cobalt, nickel, rare earths, and the like.

The relative sizes of the initial and subsequent catalytic elements may be such that their volume ratio, i.e., the superficial volume of the subsequent catalyst to the initial catlyst, including void spaces within the catalytic masses, is often at least about ... \* 72 PAGES 1431 L

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11/22/97

A31

Attachment B

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LEVEL 1 - 1 OF 4 PATENTS

5,344,815

<=2> GET 1st DRAWING SHEET OF 4

Sep. 6, 1994

Fabrication of high T C superconducting helical resonator

INVENTOR: Su, Sophia R., Weston, Massachusetts O'Connor, Margaret, Worcester, Massachusetts Butler, Scott, N. Oxford, Massachusetts

... [\*13] oxygen for at least 2 hr.

14. A method in accordance with claim 11 wherein said mixture further comprises at least about 3 w/o grain aligned clusters of a like rare earth barium copper oxide superconductor.

15. A method in accordance with claim 11 wherein said rare earth [\*15]

The state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the s

# [\*16] 16. A LEVEL 1 - 2 OF 4 PATENTS

and the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of the state of t 5,236,091

#### <=2> GET 1st DRAWING SHEET OF 5

Aug. 17, 1993

Eddy current separator and method of making a rotor

INVENTOR: Kauppila, Raymond, Marquette, Michigan Nowak, Gregory, Girard, Pennsylvania

... as follows:

[\*1] 1. A rotor for an eddy current separator comprising a rotor body having generally cylindrical, outer peripheral surfaces designed to be rotated at a design speed;

plate-like rare earth permanent magnets;

adhesive means attaching said plate-like rare earth permanent magnets to said outer peripheral surfaces of said rotor body at a bond line;

said plate-like rare earth permanent magnets being disposed in longitudinal rows extending from one end of said rotor to the other;

said plate-like rare earth permanent magnets in a particular row having a polarity on their outer end opposite the polarity of an outer end of said plate-like permanent magnets in adjacent rows;

- a fiber means ...
- ... [\*3] equal to that of carbon.
- [\*4] 4. A rotor for an eddy current separator comprising a rotor body having generally cylindrical, outer peripheral surfaces designed to be rotated at a design speed;

plate-like rare earth permanent magnets;

adhesive means attaching said plate-like rare earth permanent magnets to said outer peripheral surfaces of said rotor body at a bond line;

said plate-like rare earth permanent magnets being disposed in longitudinal rows extending from one end of said rotor to the other;

said plate-like rare earth permanent magnets in a particular row having a polarity on their outer end opposite the polarity of an outer end of said plate-like permanent magnets in adjacent rows;

fiber means wrapped ...

... [\*7] body having a polygonal outer periphery;

said polygonal outer periphery having a plurality of circumferentially disposed adjacent flat surfaces of equal width extending longitudinally of Pat. No. 5236091, \*7

said rotor from end to end thereof;

plate-like rare earth permanent magnets having a width substantially equal to the width of sides of said polygonal outer periphery and attached to said flat

surfaces by adhesive;

said plate-like rare earth permanent magnets extending substantially continuously from end to end of said rotor:

to receive said rotor;

a heat shield being ...

LEVEL 1 - 3 OF 4 PATENTS

5,162,298

<=2> GET 1st DRAWING SHEET OF 5

Nov. 10, 1992

Grain boundary junction devices using high T c superconductors

INVENTOR: Chaudhari, Praveen, Briarcliff Manor, New York
Chi, Cheng-Chung J., Yorktown Heights, New York
Dimos, Duane B., Upper Montclair, New Jersey
Mannhart, Jochen D., Metzingen, New York, Federal Republic of Germany
Tsuei, Chang C., Chappaqua, New York

- $\dots$  [\*4] copper oxide material having a superconducting onset temperature greater than 77 K.
- [\*5] 5. The device of claim 4, where said superconducting material includes an atom selected from the group consisting of rare earth atoms and rare earth-like atoms.
- [\*6] 6. The device of claim 4, where said superconducting material includes an alkaline earth atoms.
- [\*7] 7. The device of claim 4, where said superconducting material includes bismuth.
  - [\*8] 8. The device of claim 1, where ... LEVEL 1 4 OF 4 PATENTS

4,681,625

<=2> GET 1st DRAWING SHEET OF 11

Jul. 21, 1987

Methods for simultaneously desulfurizing and degassing steels

INVENTOR: Wilson, William G., 820 Harden Dr., Pittsburgh, Pennsylvania 15229

- ... [\*21] metals to be added in the tube to enhance desulfurization are those which are known to have the ability to reduce the oxygen content of the steel, but also have the ability to form sulfides which would float out of the steel into the slag which include magnesium, calcium, barium, rare earths and the like.
- [\*22] 22. The method as claimed in claims 1 or 5 wherein the ferro-alloys and elemental metals to be added in the tube are those necessary to obtain the desired chemical analysis of the finished steel such as ferro- ... \* 5 PAGES 99 LINES JOB 53156 100G6J
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Attachment C

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5,686,394

Nov. 11, 1997

Process for manufacturing a superconducting composite

INVENTOR: Sibata, Kenichiro, Hyogo, Japan Sasaki, Nobuyuki, Hyogo, Japan Yazu, Shuji, Hyogo, Japan Jodai, Tetsuji, Hyogo, Japan

SUM:

... Ho-Cu-O system or Ba-Dy-Cu-O system compound oxide which possess the quasi-perovskite type crystal structure including an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

The abovementioned type superconductors can be prepared from a powder mixture Consisting of oxides and/or carbonates containing constituent elements of said superconductor. The powder mixture may include optionally oxides and/or carbonates of at least ...

FOCUS - 2 OF 107 PATENTS

5,679,980

<=2> GET 1st DRAWING SHEET OF 5

Oct. 21, 1997

Conductive exotic-nitride barrier layer for high-dielectric-constant material electrodes

INVENTOR: Summerfelt, Scott R., Dallas, Texas

**DETDESC:** 

TABLE

\* Conductive perovskite like FOCUS - 3 OF 107 PATENTS

5,665,628

<=2> GET 1st DRAWING SHEET OF 5

Sep. 9, 1997

Method of forming conductive amorphous-nitride barrier layer for high-dielectric-constant material electrodes

INVENTOR: Summerfelt, Scott R., Dallas, Texas

DETDESC:

\*

**TABLE** 

Conductive perovskite like FOCUS - 4 OF 107 PATENTS

5,661,112

<=2> GET 1st DRAWING SHEET OF 3

Aug. 26, 1997

Superconductor

INVENTOR: Hatta, Shinichiro, 201-1028, Higashinakafuri-2-chome, Hirakata-shi, Japan Higashino, Hidetaka, A2-505, 117, Hitotsuyacho, Matsubara-shi, Japan Hirochi, Kumiko, 22, Keihanhondori-1-chome, Moriguchi-shi, Japan Adachi, Hideaki, 3-1-505, Mitsuiminamimachi, Neyagawa-shi, Japan

- ... [\*1] film being a transition metal element selected from Pt, Au, Ag, Pd, Ni and Ti the composition A-B-Cu-O of said oxide film being in the form of layered perovskite-like structure.
- [\*2] 2. A superconductor according to claim 1, wherein an additional metal film is formed on said oxide film, or the oxide films and metal films are laminated alternately to form a multi-layer structure.

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## <=2> GET 1st DRAWING SHEET OF 4

Jul. 15, 1997

Chemical vapor deposition process for fabricating layered superlattice materials

INVENTOR: Paz De Araujo, Carlos A., Colorado Springs, Colorado Watanabe, Hitoshi, Tokyo, Japan Scott, Michael C., Colorado Springs, Colorado Mihara, Takashi, Saitama, Japan

#### DETDESC

... Layered superlattice materials may be summarized more generally under the formula: [See Original Patent for Chemical Structure Diagram]

where A1, A2 . . . A represent A-site elements in the perovskite-like structure, which may be elements such as strontium, calcium, barium, bismuth, lead, and others, S1, S2 . . . Sk represent super-lattice generator elements, which usually is bismuth, but can also be materials such as yttrium, scandium, lanthanum, antimony, chromium, thallium, and other elements with a valence of + 3, B1, B2 . . . BI represent B-site elements in the perovskite-like structure, which may be elements such as titanium, tantalum, hafnium, tungsten, niobium, zirconium, and other elements, and Q represents an anion, which may be elements such as oxygen, fluorine, chlorine and hybrids of these elements, such . . .

... [\*14] s2 > ... Sk[xk] < + sk > B1[y1] < + b1 > B2[y2] < + b2 > ... B1[y1] < + b1 > Q[z] < - 2 > , where A1, A2 ... Aj represent A-site elements in a perovskite-like structure, S1, S2 ... Sk represent superlattice generator elements, B1, B2 ... B1 represent B-site elements in said perovskite-like structure, Q represents an anion, the superscripts indicate valences of the respective elements, the subscripts indicate an average number of atoms of the element in the unit cell, and at least w1 and y1 are non-zero, and wherein said A- ...

FOCUS - 6 OF 107 PATENTS

5,647,904

<=2> GET 1st DRAWING SHEET OF 2

Jul. 15, 1997

Method for manufacturing superconducting ceramics in a magnetic field

INVENTOR: Yamazaki, Shunpei, Tokyo, Japan

#### STIM .

... 300 K. by a method in which a mixture of chemicals in a suitable composition is compacted and fired. These superconducting ceramics form a quasi-molecular atomic unit in a perovskite-like structure whose unit cell is constructed with one layer in which electrons have essentially one-dimensional motion, whereas a number of crystalline grains are arranged at randam with diverse crystalline directions, and therefore the critical current density is

... cm from conventional several millimeters. The breadth and thickness may be more flexibly controlled by skilled persons according to the invention in comparison with the prior art technique.

Superconducting materials are constructed in perovskite-like structures as illustrated in FIG. 1 in accordance with the present invention. The structure comprises copper atoms 2, an intervening copper atom 3, oxygen atoms 5 and 6 surrounding the copper ...

DRWDESC:

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a schematic diagram showing the configuration of the perovskite-like molecular sturcture in accordance with the present invention.

FIGS. 2(A) and 2(B) are top and side sectional views showing an apparatus for manufacturing superconducting ceramics in accordance with the present invention.

FOCUS - 7 OF 107 PATENTS

5,646,094

<=2> GET 1st DRAWING SHEET OF 4

Jul. 8, 1997

Rare earth substituted thallium-based superconductors

INVENTOR: Tallon, Jeffrey Lewis, 3 Marine Drive, York Bay, Eastbourne, New Zealand
Presland, Murray Robert, 4/1 Mahina Bay Road, Mahina Bay, Eastbourne, New Zealand

## ABST:

... lanthanide rare earth elements and where 0.3 </= a,b </= 0.7, 0.05 </= c </= 1.1,2 - c </= d </= 1.95, 0.05 </= e </= 1, 1.9 </= f </= 2.1 and 6.5 </= g </= 7.5. These compounds, which are layered perovskite-like oxides, exhibit a high chemical stability, form readily into nearly single phase, do not require adjustment of oxygen stoichiometry after synthesis and compositions may be chosen allowing superconductivity at temperatures ...

#### SIIM

... for example, do not require adjustment of oxygen stoichiometry after synthesis, and compositions may be chosen allowing superconductivity at temperatures exceeding 100 K.

The novel compounds described herein have the same tetragonal layered perovskite-like structure of the parent compound Tl0.5Pb0.5CaSr2Cu2O7 comprising in sequence: a Tl0.5Pb0.5O layer with Tl/Pb occupying square comer-shared sites and oxygen distributed about the face centre; a SrO layer with ...

FOCUS - 8 OF 107 PATENTS

5,626,906

<=2> GET 1st DRAWING SHEET OF 3

May 6, 1997

Electrodes comprising conductive perovskite-seed layers for perovskite dielectrics

INVENTOR: Summerfelt, Scott R., Dallas, Texas Beratan, Howard R., Dallas, Texas

#### ABST:

... layer and the conductive oxide layer each comprise the same metal. The metal should be conductive in its metallic state and should remain conductive when partially or fully oxidized. Generally, the perovskite-seed layer has a perovskite or perovskite-like crystal structure and lattice parameters which are similar to the perovskite dielectric layer formed thereon. At a given deposition temperature, the crystal quality and other properties of the perovskite dielectric will generally be enhanced by depositing it on ...

SUM:

constant greater than about 50 at device operating temperature. As used herein the term "perovskite" means a material with a perovskite or perovskite-like

crystal structure. As used herein the term "dielectric", when used in reference to a perovskite, means a non-conductive perovskite, pyroelectric, ferroelectric, or high-dielectric-constant oxide material. The deposition of a ...

... structure. To facilitate perovskite crystal formation, perovskite dielectrics such as PZT have been deposited on some conductive perovskites such as YBa2Cu3O[7 - x ]and (La,Sr)CoO3. Deposition of PZT on a substrate with a perovskite or perovskite-like crystal structure normally minimizes the formation of the pyrochlore phase and improves the properties of the perovskite dielectric. However, the materials used thus far for the deposition surface have several problems. For example, they typically involve new cations such ...

... layer each comprise the same metal. The metal should be conductive in its metallic state and should remain conductive when partially or fully oxidized, and when in a perovskite. Generally, the perovskite-seed layer has a perovskite or perovskite-like crystal structure and lattice parameters which are similar to the perovskite dielectric layer formed thereon. At a given deposition temperature, the crystal quality and other properties of the perovskite dielectric will generally be enhanced by depositing it on ...

**DETDESC:** 

TABLE

ruthenate

seed layer perovskite-like materials FOCUS - 9 OF 107 PATENTS

5,611,854

Mar. 18, 1997

Seed crystals with improved properties for melt processing superconductors for practical applications

INVENTOR: Veal, Boyd W., Downers Grove, Illinois Paulikas, Arvydas, Downers Grove, Illinois Balachandran, Uthamalingam, Hinsdale, Illinois Zhong, Wei, Chicago, Illinois

## DETDESC:

... Although PbTiO3 is shown in the Table, other perovskites of the form RTiO3, when R is La or a rare earth are good candidates. EuTiO3 has a lattice parameter of 3,897 [Angstrom] . NdGaO3, and other perovskite-like oxides with the prototype GdFeO3 structure should also serve well. NdGaO3 is available as a commercial substrate material. Others may also be commercially available, particularly LaCrO3 which has many industrial applications.

Oxides with the GdFeO3 ( ...

FOCUS - 10 OF 107 PATENTS

5,602,080

<=2> GET 1st DRAWING SHEET OF 1

Feb. 11, 1997

Method for manufacturing lattice-matched substrates for high-T[c] superconductor films

INVENTOR: Bednorz, Johannes G., Wolfhausen, Switzerland Mannhart, Jochen D., Thalwil, Switzerland Mueller, Carl A., Hedingen, Switzerland Schlom, Darrell G., State College, Pennsylvania

... a close match-preferably approaching an ideal match-of the lattice parameters of a substrate-without a buffer layer-to a selected high-T[c

]superconductor material having a perovskite or a perovskite-like crystal structure can be achieved by a method comprising the following steps:

Determining the relevant lattice constant or constants of the selected superconductor material; choosing a desired orientation of the superconductor layer to ...

... for the deposition of the superconductor.

One preferred method of the invention for manufacturing a lattice-matched substrate for a film of a selected high-T[c] superconductor material having a perovskite or perovskite-like crystal structure at a selected orientation relative to the film dimensions comprises the steps set forth below.

The preferred method of the invention includes the step of determining a relevant lattice constant or constants of the selected ...

... make the codeposition from separate sources each containing one or more of the materials combined to form the buffer layer.

Preferred substrate component materials include strontium titanate SrTiO3 and lanthanum aluminate LaAlO3 for perovskite-like superconductor materials such as YBa2Cu3O7 - delta .

In the following description, a preferred method for manufacturing crystalline substrate material having essentially the same lattice constant as the corresponding lattice constant of a ...

FOCUS - 11 OF 107 PATENTS

5,593,951

<=2> GET 1st DRAWING SHEET OF 4

Jan. 14, 1997

Epitaxy of high T[C] superconductors on silicon

INVENTOR: Himpsel, Franz J., Mt. Kisco, New York

SUM:

... first showed superconducting behavior in mixed copper-oxides, typically including rare earth and/or rare earth-like elements and alkaline earth elements, for example La, Ba, Sr, . . . , and having a perovskite-like structure.

Materials including the so called "1-2-3" phase in the Y-Ba-Cu-O system have been found to exhibit a superconducting transition temperature in excess of 77K. R. B. ...

FOCUS - 12 OF 107 PATENTS

5,590,053

<=2> GET 1st DRAWING SHEET OF 20

Dec. 31, 1996

Method of determining a space group

INVENTOR: Ito, Tatsuya, Kawasaki, Japan Kawai, Masahito, Kawasaki, Japan Yasukawa, Yoshihito, Kawasaki, Japan

## DETDESC:

... present invention will be described with reference to FIG. 15 to FIG. 20.

Let it be assumed here that a crystal as a target of analysis is one of

LaGdSrCuO4. In the case of investigation into such a perovskite like copper

oxide superconductor, it is an effective technique of investigating a new
substance to laminate partial structures to grasp a laminate structure

ا المستقدم المنظم ا المنظم المنظم المنظم المنظم المنظم المنظم المنظم المنظم المنظم المنظم المنظم المنظم المنظم المنظم المنظم المنظ characteristic of the substance. The structure analysis of the target crystal by this technique will ...

FOCUS - 13 OF 107 PATENTS

5,589,284

<=2> GET 1st DRAWING SHEET OF 3

Dec. 31, 1996

Electrodes comprising conductive perovskite-seed layers for perovskite dielectrics

INVENTOR: Summerfelt, Scott R., Dallas, Texas Beratan, Howard R., Dallas, Texas

## ABST:

... layer and the conductive oxide layer each comprise the same metal. The metal should be conductive in its metallic state and should remain conductive when partially or fully oxidized. Generally, the perovskite-seed layer has a perovskite or perovskite-like crystal structure and lattice parameters which are similar to the perovskite dielectric layer formed thereon. At a given deposition temperature, the crystal quality and other properties of the perovskite dielectric will generally be enhanced by depositing it on ...

## SUM:

... As used herein, the term "high-dielectric-constant" means a dielectric constant greater than about 50 at device operating temperature. As used herein the term "perovskite" means a material with a perovskite or perovskite-like crystal structure. As used herein the term "dielectric", when used in reference to a perovskite, means a non-conductive perovskite, pyroelectric, ferroelectric, or high-dielectric-constant oxide material. The deposition of a ...

... structure. To facilitate perovskite crystal formation, perovskite dielectrics such as PZT have been deposited on some conductive perovskite such as YBa2Cu3O[7-x ]and (La,Sr)CoO3. Deposition of PZT on a substrate with a perovskite or perovskite-like crystal structure normally minimizes the formation of the pyrochlore phase and improves the properties of the perovskite dielectric. However, the materials used thus far for the deposition surface have several problems. For example, they typically involve new cations such ...

... layer each comprise the same metal. The metal should be conductive in its metallic state and should remain conductive when partially or fully oxidized, and when in a perovskite. Generally, the perovskite-seed layer has a perovskite or perovskite-like crystal structure and lattice parameters which are similar to the perovskite dielectric layer formed thereon. At a given deposition temperature, the crystal quality and other properties of the perovskite dielectric will generally be enhanced by depositing it on ...

**DETDESC:** 

TABLE

ruthenate

seed layer perovskites or perovskite
\* like materials (e.g.

FOCUS - 14 OF 107 PATENTS

5,585,300

<=2> GET 1st DRAWING SHEET OF 5

Dec. 17, 1996

Method of making conductive amorphous-nitride barrier layer

for-high-dielectric-constant material electrodes

INVENTOR: Summerfelt, Scott R., Dallas, Texas

DETDESC:

TABLE

Conductive perovskite like
FOCUS - 15 OF 107 PATENTS

5,583,096

<=2> GET 1st DRAWING SHEET OF 8

Dec. 10, 1996

Superconductive compounds and process for producing said compounds

INVENTOR: Cavazos, Ramon G., Paseo de la Reforma 403, Primer Piso, Mexico D.F. 06500

#### DETDESC:

... A. Muller in their article entitled "Possible High Tc Superconductivity in Ba-La-Cu-O System". (Zeitschrift fur Physik B-Condensed Matter 64,189-193 (1986), reported: "... perovskite-like-mixed valent copper compound. Upon cooling, the samples show a linear decrease in resistivity, then an approximately logarithmic increase, interpreted as a beginning of localization. Finally, an abrupt decrease by ...

FOCUS - 16 OF 107 PATENTS

5,563,331

<=2> GET 1st DRAWING SHEET OF 3

Oct. 8, 1996

Magnetoresistive sensor utilizing a sensor material with a perovskite-like crystal structure

INVENTOR: Von Helmolt, Rittmar, Erlangen, Federal Republic of Germany Wecker, Joachim, Roettenbach, Federal Republic of Germany

## ABST:

A magnetoresistive sensor may be constructed with material having a perovskite-like crystal structure and an increased magnetoresistive effect. The material is based on the composition (A1)[1-x](A2)[x]MnO[z], with A1 (trivalent) selected from Y, La, or a lanthanide, A2 (bivalent) from an alkaline-...

### SUM:

## BACKGROUND OF THE INVENTION

The present invention relates to a magnetoresistive sensor with a layer made of a sensor material that possesses a perovskite-like crystal structure and exhibits an increased magnetoresistive effect.

The general structure and operation of magnetoresistive sensors with thin films made of ferromagnetic transition metals are explained further in, for example, the book "Sensors", Vol. ...

... x]Se (cf. "Journal of Applied Physics," Vol. 38, No. 3, Mar. 1, 1967, pp. 959-964). A corresponding effect is also evident in Nd0.5Pb0.5Mn03 crystals; these crystals have a perovskite-like structure (cf. "Physics B," Vol. 155, 1989, pp. 362-365). However, the change in electrical resistance as a function of magnetic induction observed in these material systems is confined to low ...

... occur only to a reduced extent, in a sensor material that is the subject of a German patent application No. P 43 10 318.9 (not previously disclosed).

This material possesses a perovskite-like crystal structure and exhibits an increased magnetoresistive effect. A composition based on (A1)[1-x](A2)[x]MnO[x lis to be selected for the material, such that the trivalent constituent Al at least contains ...

... sensor according to an embodiment of the present invention includes at least two layers, a first layer and a second layer. Each of the first and second layers is made of a sensor material that possesses a perovskite-like crystal structure and exhibits an increased magnetoresistive effect. The sensor material of each of the first and second layers has a composition based on (A1)[1-x](A2)[x]MnO[z], where Al is a trivalent ...

... indicated can also contain minimal impurities with less than 0.5 atomic percent of each impurity element. Exemplary embodiments for corresponding materials are therefore La0.67Ba0.33Mn03, or Pr0.5Sr0.5Mn03, or Nd0.33Ca0.67Mn03, or (Dy0.67Mg0.33)(Mn0.8Cu0.2)02.9. All these materials have Pat. No. 5563331, \*

FOCUS

a perovskite-like crystal structure and are characterized by an increased magnetoresistive effect M[r ]of, in particular, more than 10%, and preferably more than 50%. The effect is thus considerably greater than in known Cu/Co multilayer systems.

- ... 1557-1559). According to the present invention, corresponding layers of the sensor material are advantageously deposited onto substrates whose respective crystalline unit cell has dimensions matched to the unit cell of the sensor material. Substrate materials that also have a perovskite-like crystal structure are therefore particularly suitable. Corresponding exemplary embodiments are SrTiO3, MgO, LaA1O3, NdGaO3, MgA12O4, or Y-stabilized ZrO2 (abbreviated YSZ). In addition, however, Si substrates that are coated with a special intermediate ...
  - a layer system comprising at least two layers, including: ... [\*1]
  - a first layer; and
  - a second layer;

wherein each of said first and second layers comprises a sensor material that possesses a perovskite-like crystal structure and exhibits an increased magnetoresistive effect, such that the sensor material of each of said first and second layers has a composition based on (A1)[1-x](A2)[x]MnO[z], wherein A1 is a

- similar to said first layer and layers similar to said second ... [\*4] layer.
- 5. A magnetoresistive sensor according to claim 2; wherein the layer system is deposited on a substrate made of a material that has a perovskite-like crystal structure.
- 6. A magnetoresistive sensor according to claim 1, wherein the first layer and the second layer have different thicknesses.
- 7. A magnetoresistive sensor according to claim 6, wherein the layer system includes ...
- ... [\*7] similar to said first layer and layers similar to said second

layer.

[28] 8. A magnetoresistive sensor according to claim 6, wherein the layer. system is deposited on a substrate made of a material that has a perovskite-like crystal structure.

- [#9] 9. A magnetoresistive sensor according to claim 1, wherein the layer system includes more than two layers which alternate between layers similar to said first layer and layers similar to said second layer.
- 10. A magnetoresistive sensor according to claim 9, wherein the layer system is deposited on a substrate made of a material that has a Pat. No. 5563331, \*10

**FOCUS** 

perovskite-like crystal structure.

- 11. A magnetoresistive sensor according to claim 1, wherein the layer system is deposited on a substrate made of a material that has a perovskite-like crystal structure.
- 12. A magnetoresistive sensor according to claim 1, wherein 0.25 </= [\*12] x < /= 0.75.
  - [\*13] 13. A magnetoresistive sensor according to claim 1, wherein z = 3. FOCUS - 17 OF 107 PATENTS

5,554,585

<=2> GET 1st DRAWING SHEET OF 1

Sep. 10, 1996

Method of forming a superconductor microstrip transmission line

INVENTOR: Simon, Randy W., Long Beach, California Platt, Christine E., El Segundo, California Lee, Alfred E., Torrance, California Lee, Gregory S., West Los Angeles, California

## REF-CITED:

... 61(1):28-35 (1973).

Geballe, "Paths to Higher Temperature Superconductors," Science, vol. 259, Mar.

12, 1993, pp. 1550-1551.

Geller, S., et al., "Crystallographic Studies of Perovskite-like Compounds. II.

"Arts Count 9:1019-1025 (1956).

Rare Earth Aluminates," Acta. Cryst., 9:1019-1025 (1956). Geller, S., "Crystallographic Studies of Perovskite-like Compounds. IV. Rare Earth Scandates, Vanadites, Galliates, Orthochromites," Acta Cryst., 10:243-248 (1957).

Gulyaev,, Yu V., et al., "YBa2Cu3O[7 - x ]Films with a High-temperature ... FOCUS - 18 OF 107 PATENTS

5,552,373

<=2> GET 1st DRAWING SHEET OF 2

Sep. 3, 1996

Josephson junction device comprising high critical temperature crystalline copper superconductive layers

INVENTOR: Agostinelli, John A., Rochester, New York Mir, Jose M., Webster, New York Lubberts, Gerrit, Penfield, New York Chen, Samuel, Penfield, New York

## **DETDESC:**

... can take any convenient form capable of permitting deposition of USCO" CARREST COMMENTS OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPERTY OF THE PROPER thereon as a thin film.

mani in departe. 1 d'air.

In a specifically preferred form of the invention SUB" is chosen from

materials that themselves exhibit a perovskite or perovskite-like crystal structure. Strontium titanate is an example of a perovskite crystal structure which is specifically preferred for use as a substrate. Lanthanum aluminate (LaA103), lanthanum gallium oxide (LaGa03) and potassium tantalate are ... FOCUS - 19 OF 107 PATENTS

5,527,567

<=2> GET 1st DRAWING SHEET OF 6

Jun. 18, 1996

Metalorganic chemical vapor deposition of layered structure oxides

INVENTOR: Desu, Seshu B., Blacksburg, Virginia Tao, Wei, Blacksburg, Virginia Peng, Chien H., Blacksburg, Virginia Li, Tingkai, Blacksburg, Virginia Zhu, Yongfei, Blacksburg, Virginia

SUM:

... 1961), 695; G. A. Smolenski, V. A. Isupov and A. I. Agranovskaya, Fiz Tverdogo Tela, 3, (1961), 895). These compounds have a pseudo-tetragonal symmetry and the structure is comprised of stacking of perovskite-like units between (Bi2O2)<2 + > layers along the pseudo-tetragonal c-axis. A large number of these compounds do not contain any volatile components in their sublattice that exhibits spontaneous polarization. The tendency for ...

> PAGE 22

FOCUS - 20 OF 107 PATENTS

5,523,283

<=2> GET 1st DRAWING SHEET OF 1

Jun. 4, 1996

L[a]AlO3 Substrate for copper oxide superconductors

INVENTOR: Simon, Randy W., Long Beach, California Platt, Christine E., El Segundo, California Lee, Alfred E., Torrance, California Lee, Gregory S., West Los Angeles, California

REF-CITED:

... 61(1):28-35 (1973).

Gaballe, "Paths to Higher Temperature Superconductors," Science, vol. 259, Mar. 12, 1993, pp. 1550-1551.

Geller, S., et al., "Crystallographic Studies of Perovskite-like Compounds. II. Rare Earth Aluminates," Acta. Cryst., 9:1019-1025 (1956).

Geller, S., "Crystallographic Studies of Perovskite-like Compounds. IV. Rare Earth Scandates, Vanadites, Galliates, Orthochromites," Acta Cryst., 10:243-428

Gulysev, Yu V., et al., "YBa2CU30[7-x ]Films with a High-temperature ... FOCUS - 21 OF 107 PATENTS

5,523,282

<=2> GET 1st DRAWING SHEET OF 1

Jun. 4, 1996

High-frequency substrate material for thin frim layered perovskite supertonductors

INVENTOR: Simon, Randy W., Long Beach, California

Platt, Christine E., Er Segundo, California Lee, Alfred E., Torrance, California Lee, Gregory S., West Los Angeles, California

## REF-CITED:

... A., et al., "The Flux Shuttle-A Josephson Junction Shift Register Employing Single Flux Quanta," Proceedings of the IEEE, 61(1):28-35 (1973). Geller, S., "Crystallographic Studies of Perovskite-like Compounds. Rare Earth Scandates, Vanadites, Galliates, Orthochromites," Acta Cryst., 10:243-251 (1957).

Gurvitch, M., et al., "Preparation and Substrate Reactions of Superconducting Y-Ba-Cu-O Films,"  $\dots$ 

... in the Coprecipitation of Carbonate and Hydroxide Compounds of Lanthanum and Aluminum," Russian Journal of Inorganic Chemistry, vol. 22, No. 11, pp. 1622-1625, 1977.

S. Geller et al., "Crystallographic Studies of Perovskite-like Compounds. II. Rare Earth Aluminates," Acta Cryst., vol. 9, pp. 1019-1025, 1956.

J. Kilner et al., "Electrolytes for the High Temperature Fuel Cell; Experimental and Theoretical ...

FOCUS - 22 OF 107 PATENTS

5,519,234

<=2> GET 1st DRAWING SHEET OF 30

May 21, 1996

Ferroelectric dielectric memory cell can switch at least giga cycles and has low fatigue - has high dielectric constant and low leakage current

INVENTOR: Paz de Araujo, Carlos A., Colorado Springs, Colorado Cuchiaro, Joseph D., Colorado Springs, Colorado Scott, Michael C., Colorado Springs, Colorado McMillan, Larry D., Colorado Springs, Colorado

#### ARCT .

...  $s2 > \ldots$  Sk xk < + ak > B1 yl < + bl> B2 y2 < + b2  $> \ldots$  B1 yl < + bl> Q z < - 2> , where A1, A2 ... Aj represent A-site elements in a perovskite-like structure, S1, S2 ... Sk represent superlattice generator elements, B1, B2 ... B1 represent B-site elements in a perovskite-like structure, Q represents an anion, the superscripts indicate the valences of the respective elements, the subscripts indicate the number of atoms of the element in the unit cell, and at least w1 and y1 are non-zero. Some of these materials are extremely low ...

#### STIM .

- ... 676 (1962) and Chapter 8 pages 241-292 and pages 624 & 625 of Appendix F of the Lines and Glass reference cited above. As outlined in section 15.3 of the Smolenskii book, the layered perovskite-like materials can be classified under three general types:
- (I) compounds having the formula A m 1 Bi2M m O 3m + 3 , where A = Bi<3 + > , Ba<2 + > , Sr< ...
  - ... strontium titanates Sr2TiO4, Sr3Ti2O7 and Sr4Ti3O1O; and
- (III) compounds having the formula A m M m O 3m + 2, including compounds such as Sr2Nb2O7, La2Ti2O7, Sr5TiNb4O17, and Sr6Ti2Nb4O2O.

Smolenskii pointed out that the perovskite-like layers may have different thicknesses, depending on the value of m, and that the perovskite AMO3 is in principal the limiting example of any type of layered perovskite-like structure with m = infinity. Smolenskii also noted that if the layer with minimum thickness (m = 1) is denoted by P and the bismuth-oxygen layer is denoted by B, then the type I compounds may be described as . . . BP m BP m . . . . Further

Smolenskii noted that if m is a fractional number then are lattice contains perovskite-like layers of various thicknesses, and that all the known type I compounds are ferroelectrics. Similarly, Smolenskii noted that the type two compounds could be represented as . . . SP m SP m . . . where P is the perovskite-like layer of thickness m and S is the strontium-oxygen connecting layer, and that since the type I and type II compounds have similar perovskite-like layers, the existence of "hybrid" compounds such as . . . BP m SP n BP m SP m . . . "should not be ruled out", though none had been obtained at that time.

Pat. No. 5519234, \*

**FOCUS** 

Up to now, these layered ferroelectric ...

...  $s2 > \ldots$  Sk xk < + sk > B1 y1 < + b1 > B2  $y2 < + b2 > \ldots$  Bl y1 < + b1 > Q z < - 2 >, where A1, A2 ... Aj represent A-site elements in a perovskite-like structure, S1, S2 ... Sk represent superlattice generator elements, B1, B2 ... Bl represent B-site elements in a perovskite-like structure, Q represents an anion, the superscripts indicate the valences of the respective elements, the subscripts indicate the average number of atoms of the element in the unit cell, and at least w1 and y1 are non-zero. Preferably, the A-

... layered superlattice material comprises a material having a localized structure, within a grain or other larger or smaller unit, which localized structure contains predominately repeatable units containing one or more perovskite-like layers and one or more intermediate non-perovskite-like layers spontaneously linked in an interdependent manner.

In another aspect the invention provides a non-volatile ferroelectric memory comprising: a ferroelectric memory cell including a layered superlattice ...

#### **DETDESC:**

... curves as shown in FIG. 5C, which show fatigue of less than 30%, which is much less than for any ferroelectric material on which endurance tests had been performed in the prior art. It was realized that the SrBi4Ti4O15 was one of the layered perovskite-like materials catalogued by Smolenskii, and thought that perhaps the natural layered structure of these materials might be the source of the low-fatigue property. Other devices were fabricated having the structure shown in FIG. 2C, i.e. a ...

13, a layered superlattice material 92 is illustrated. Smolenskii recognized that what we call the layered superlattice materials spontaneously form into layers 94 with a perovskite-like structure which alternate with layers 96 having a simpler structure. Depending on the material, the perovskite-like layers 94 may include one or a plurality of linked layers of perovskite-like octahedrons 90. As an example, FIG. 14 shows a unit cell of the material ABi2B2< + 5> 09, which is the formula for strontium bismuth tantalate (SrBi2Ta2O9) and other layered superlattice materials, such as tantalum, niobium, and tungsten, having a element with a valence of + 5 in the B-site. In this structure, each perovskite-like layer 94 includes two layers of octahedrons 90 which are separated by layers 96 of a material that does not have a perovskite-like structure. In this material the primitive unit cell consists of two perovskite layers 94 and two non-perovskite layers 96, since the structure shifts between the layers 98A and 98B. In FIG. ...

... 015, which is the formula for strontium bismuth titanate (SrBi4Ti4015) and other layered superlattice materials having an element, such as titanium, hafnium, and zirconium, having a valence of + 4 in the B-sites. In this material each the perovskite-like layer 94 has four layers of octahedrons 90.

As the understanding of what Smolenskii called a layered perovskite-like structure increased, the inventors have realized that these materials are more than a substance which spontaneously forms in layers. This is seen most easily by an example. Strontium bismuth tantalate (SrBi2Ta2O9) can be considered to Pat. No. 5519234, \*

be ...

... in the following definition: (B) a material having a localized structure, within a grain or other larger or smaller unit, which localized structure contains predominately repeatable units containing one or more perovskite-like layers and one or more intermediate non-perovskite-like layers spontaneously linked in an interdependent manner.

It has been discovered that the layered superlattice materials catalogued by Smolenskii et al. are all likely candidates for fatigue free switching ferroelectrics and dielectric materials that are resistant to ...

... x2 < + s2 > . . . Sk xk < + sk > B1 y1 < + b1 > B2 y2 < + b2 > . . . B1 y1 < + b1 > Q z < - 2 > ,

where A1, A2 . . . Aj represent A-site elements in the perovskite-like structure, which may be elements such as strontium, calcium, barium, bismuth, lead, and others S1, S2 . . . Sk represent superlattice generator elements, which usually is bismuth, but can also be materials such as yttrium, scandium, lanthanum, antimony, chromium, thallium, and other elements with a valence of + 3, B1, B2 . . . Bl represent B-site elements in the perovskite-like structure, which may be elements such as titanium, tantalum, hafnium, tungsten, niobium, zirconium, and other elements, and Q represents an anion, which generally is oxygen but may also be other elements, such as fluorine, ...

... [ $\pm$ 2] s2 > . . . Sk xk < + sk> B1 y1 < + b1> B2 y2 < + b2 > . . . B1 y1 < + b1> Q z < - 2> , where A1, A2 . . . Aj represent A-site elements in a perovskite-like structure, S1, S2 . . . Sk represent superlattice generator elements, B1, B2 . . . B1 represent B-site elements in a perovskite-like structure, Q represents an anion, the superscripts indicate the valences of the respective elements, the subscripts indicate the average number of atoms of the element in the unit cell, and at least w1 and y1 are non-zero.

[\*3] 3. A ...

FOCUS - 23 OF 107 PATENTS

5,504,041

<=2> GET 1st DRAWING SHEET OF 5

Apr. 2, 1996

Conductive exotic-nitride barrier layer for high-dielectric-constant materials

INVENTOR: Summerfelt, Scott R., Dallas, Texas

**DETDESC:** 

TABLE

Conductive perovskite like materials FOCUS - 24 OF 107 PATENTS

5,489,548

<=2> GET 1st DRAWING SHEET OF 3

Feb. 6, 1996

Method of forming high-dielectric-constant-material electrodes comprising sidewall-spacers

INVENTOR: Nishioka, Yasushiro, Tsukuba, Texas, Japan Summerfelt, Scott R., Dallas, Texas

Park, Kyung-Ho, Tsukuba, Japan Bhattacharya, Pijush, Midnapur, India

DETDESC:

TABLE

# Conductive perovskite like
FOCUS - 25 OF 107 PATENTS

5,478,610

<=2> GET 1st DRAWING SHEET OF 5

Dec. 26, 1995

Metalorganic chemical vapor deposition of layered structure oxides

INVENTOR: Desu, Seshu B., Blacksburg, Virginia Tao, W., Blacksburg, Virginia

SUM:

... 34, (1961), 695; G. A. Smolenski, V. A. Isupov and A. I. Agranovskaya, Fiz Tverdogo Tela, 3, (1961), 895). These compounds have pseudo-tetragonal symmetry and the structure is comprised of stacking of perovskite-like units between (Bi202)<2 + > layers along the pseudo-tetragonal c-axis. A large number of these compounds do not contain any volatile components in their sublattice that exhibits spontaneous polarization. The tendency for ...

FOCUS - 26 OF 107 PATENTS

5,468,679

<=2> GET 1st DRAWING SHEET OF 27

Nov. 21, 1995

Process for fabricating materials for ferroelectric, high dielectric constant, and integrated circuit applications

INVENTOR: Paz de Araujo, Carlos A., Colorado Springs, Colorado Scott, Michael C., Colorado Springs, Colorado Cuchiaro, Joseph D., Colorado Springs, Colorado McMillan, Larry D., Colorado Springs, Colorado

SUM:

... 676 (1962) and Chapter 8 pages 241-292 and pages 624 & 625 of Appendix F of the Lines and Glass reference cited above.

As outlined in section 15.3 of the Smolenskii book, the layered perovskite-like materials can be classified under three general types:

- (I) compounds having the formula A m-1 Bi2M m O 3m + 3 , where A = Bi<3 + > , Ba<2 + > , Sr< ...
- ... s2> . . . Sk xk < + sk> B1 y1 < + b1> B2 y2 < + b2> . . . B1 y1 < + b1> Q z < 2> ,

**DETDESC:** 

... compatible with, or can be designed to be compatible with, the other

materials commonly used in integrated circuits, such as silicon and gallium arsenide.

The class of materials are those disclosed by Smolenskii as having a layered perovskite-like structure, as discussed in the Background of the Invention. It has been realized that these materials are more than a substance which spontaneously forms in layers. This is seen most easily by an example. Strontium bismuth tantalate (SrBi2Ta2O9) can ...

... in the following definition: (B) a material having a localized structure, within a grain or other larger or smaller unit, which localized structure contains predominately repeatable units containing one or more perovskite-like layers and one or more intermediate non-perovskite-like layers spontaneously linked in an interdependent manner.

It is well-known that compounds having the perovskite structure may be described in terms of the general formula ABQ3, where A and B are cations and Q is an anion. In the  $\dots$ 

Pat. No. 5468679, \*

**FOCUS** 

... flexible than the lattice of a ferroelectric material. Turning to FIG. 13, a layered superlattice material 92 is illustrated. Smolenskii recognized that what we call the layered superlattice materials spontaneously form into layers 94 with a perovskite-like structure which alternate with layers 96 having a simpler structure. Depending on the material, the perovskite-like layers 94 may include one or a plurality of linked layers of perovskite-like octahedrons 90. As an example, FIG. 14 shows a unit cell of the material ABi2B2< + 5> 09, which is the formula for strontium bismuth tantalate (SrBi2Ta2O9) and other layered superlattice materials, such as tantalum, niobium, and tungsten, having a element with a valence of + 5 in the B-site. In this structure, each perovskite-like layer 94 includes two layers of octahedrons 90 which are separated by layers 96 of a material that does not have a perovskite-like structure. In this material the primitive unit cell consists of two perovskite layers 94 and two non-perovskite layers 96, since the structure shifts between the layers 98A and 98B. In FIG. ...

... 015, which is the formula for strontium bismuth titanate (SrBi4Ti4O15) and other layered superlattice materials having an element, such as titanium, hafnium, and zirconium, having a valence of + 4 in the B-sites. In this material each the perovskite-like layer 94 has four layers of octahedrons 90.

It has been discovered that the layered superlattice materials catalogued by Smolenskii et al. are all likely candidates for fatigue free switching ferroelectrics and dielectric materials that are resistant to ...

... x2 < + s2 > . . . Sk xk < + sk > B1 y1 < + b1 > B2 y2 < + b2 > . . . B1 y1 < b1 > Q z < - 2 > ,

where A1, A2 . . . Aj represent A-site elements in the perovskite-like structure, which may be elements such as strontium, calcium, barium, bismuth, lead, and others S1, S2 . . . Sk represent superlattice generator elements, which usually is bismuth, but can also be materials such as yttrium, scandium, lanthanum, antimony, chromium, thallium, and other elements with a valence of + 3, B1, B2 . . . Bl represent B-site elements in the perovskite-like structure, which may be elements such as titanium, tantalum, hafnium, tungsten, niobium, zirconium, and other elements, and Q represents an anion, which generally is oxygen but may also be other elements, such as fluorine, ...

FOCUS - 27 OF 107 PATENTS

5,447,908

<=2> GET 1st DRAWING SHEET OF 1

Sep. 5, =1995

Superconducting thin film and a method for preparing the

INVENTOR: Itozaki, Hideo, Hyogo, Japan Tanaka, Saburo, Hyogo, Japan Fujita, Nobuhiko, Hyogo, Japan Yazu, Shuji, Hyogo, Japan Jodai, Tetsuji, Hyogo, Japan

#### SUM:

... structure. The term of quasi-perovskite type means a structure which can be considered to have such a crystal structure that is similar to Perovskite-type oxides and includes an orthorhombically distorted perovskite or a distorted oxygendeficient perovskite or the like.

The superconducting thin film may be also another type of superconductor consisting mainly of a compound oxide represented by the formula:

THETA 4( PHI 1-q ,Ca q ) m Cu ... FOCUS - 28 OF 107 PATENTS

5,447,906

Sep. 5, 1995

Thin film high TC oxide superconductors and vapor deposition methods for making the same

INVENTOR: Chaudhari, Praveen, Briarcliff Manor, New York Gambino, Richard J., Yorktown Heights, New York Koch, Roger H., Amawalk, New York Lacey, James A., Mahopac, New York Laibowitz, Robert B., Peekskill, New York Viggiano, Joseph M., Wappingers Falls, New York

#### SUM:

... areas.

It is another object of the present invention to provide continuous, smooth copper oxide superconductive films exhibiting superconductivity at temperatures in excess of 400 K. and methods for making these films, where the films exhibit perovskite-like structure.

It is another object of this invention to provide transition metal oxide superconductive films including a rare earth element, or rare earth-like element, where the films exhibit superconductivity at temperatures greater than 400 ...

... earth-like element, B is an alkaline earth element, and y is sufficient to satisfy valence demands of the composition.

It is another object of the present invention to provide smooth, continuous copper oxide superconducting films having a perovskite-like crystal structure and exhibiting superconductivity at temperatures in excess of 40o K., and to provide methods for making these films.

SUMMARY OF THE INVENTION

The films of this invention are oxide superconductors exhibiting superconductivity at temperatures in excess of ...

... addition to being continuous, smooth, and of excellent compositional uniformity. The Cu oxide films are therefore considered to be unique examples of this class of films, as are the processes for making them.

Typically, the films are characterized by a perovskite-like crystalline structure, such as those described in more detail by C. Michel and B. Rayeau in Revue Dde:

Chimie Minerale, 21, p. 407 (1984). These films are Formed by a ...
FOCUS - 29 OF 107 PATENTS

5,439,878

<=2> GET 1st DRAWING SHEET OF 21

Aug. 8, 1995

Method for preparing copper oxide superconductor containing carbonate radicals

INVENTOR: Kinoshita, Kyoichi, Hoya, Japan Yamada, Tomoaki, Higashimurayama, Japan

SUM:

... novel superconducting material. Description of the Prior Art

Several types of copper oxide superconductors have been discovered since high-To superconductivity was detected in the La-Ba-Cu-O system. Superconductivity would arise from the layered perovskite-like structure having CuO6 octahedra, or CuO5 pyramids, or CuO2 square planes as a building unit. The layered perovskite-like structure and a sufficient carrier concentration of the material are essential factors for making the material superconducting as indicated by Osamura & Zhang (Japan. J.Appl.Phys.26, L2094-L2096, 1987). ...

FOCUS - 30 OF 107 PATENTS

5,439,876

<=2> GET 1st DRAWING SHEET OF 5

Aug. 8, 1995

Method of making artificial layered high T c superconductors

INVENTOR: Graf, Volker, Wollerau, Switzerland Mueller, Carl A., Hedingen, Switzerland

DETDESC:

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

One material particularly suited as a substrate in the epitaxial growth of high T c superconductor material is strontium titanate, SrTiO3, which forms crystals like perovskite (FIG. 1). Each titanium ion 1 is octahedrally surrounded by six oxygen ions 2, the bigger strontium ions 3 being disposed in the spaces in between. At room temperature, ...

FOCUS - 31 OF 107 PATENTS

5,426,092

<=2> GET 1st DRAWING SHEET OF 14

Jun. 20, 1995

Continuous or semi-continuous laser ablation method for depositing fluorinated superconducting thin film having basal plane alignment of the unit cells deposited on non-lattice-matched substrates

INVENTOR: Ovshinsky, Stanford R., Bloomfield Hills, Michigan Young, Rosa, Troy, Michigan

... growth of a crystalline superconducting material in a manner as if mimicking the orientation of a substrate having an identical lattice structure

without the presence of such a substrate lattice structure. Simply stated, an "epitaxial-like" perovskite superconducting material grown on a non-lattice-matched substrate would nonetheless be characterized by a lattice structure identical to the lattice structure which would be present if the material was grown on a perovskite substrate. Thus, "...

FOCUS - 32 OF 107 PATENTS

5,424,282

<=2> GET 1st DRAWING SHEET OF 5

Jun. 13, 1995

Process for manufacturing a composite oxide superconducting wire

INVENTOR: Yamamoto, Susumu, Hyogo, Japan Murai, Teruyuki, Hyogo, Japan Kawabe, Nozomu, Hyogo, Japan Awazu, Tomoyuki, Hyogo, Japan Yazu, Shuji, Hyogo, Japan Jodai, Tetsuji, Hyogo, Japan

#### DETDESC:

... term of "quasiperovskite type structure" means any oxide that can be considered to have such a crystal structure-that is similar to perovskite-type oxides and may include an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

In practice, the element ct is preferably selected from Ba, Sr and/or Ca and the element beta is preferably selected from Y, La and/or lanthanid such as Sc, Ce, Gd, Ho, Er, Tin, Y b, ...

FOCUS - 33 OF 107 PATENTS

5,423,285

<=2> GET 1st DRAWING SHEET OF 27

Jun. 13, 1995

Process for fabricating materials for ferroelectric, high dielectric constant, and integrated circuit applications

INVENTOR: Paz de Araujo, Carlos A., Colorado Springs, Colorado Cuchiaro, Joseph D., Colorado Springs, Colorado Scott, Michael C., Colorado Springs, Colorado McMillan, Larry D., Colorado Springs, Colorado

## SUM:

... 676 (1962) and Chapter 8 pages 241-292 and pages 624& 625 of Appendix F of the Lines and Glass reference cited above.

As outlined in section 15.3 of the Smolenskii book, the layered perovskite-like materials can be classified under three general types:

- (I) compounds having the formula A m-1 Bi2M m O 3m + 3 , where A = Bi<3 + > , Ba<2 + > , Sr< ...
- ... s2> . . . Sk xk < + sk> B1 y1 < + b1> B2 y2 < + b2> . . . B1 y1 < + b1> Q z < 2> , where A1, A2 . . . Aj represent A-site elements in a perovskite-like structure, S1, S2 . . . Sk represent superlattice generator elements, B1, B2 . . . B1 represent B-site elements in a perovskite-like structure, Q represents an anion, the superscripts indicate the valences of the respective elements, the subscripts indicate the average number of atoms of the element in the unit cell, and at least w1 and y1 are non-zero. Preferably, the A-...

**DETDESC:** 

... compatible with, or can be designed to be compatible with, the other materials commonly used in integrated circuits, such as silicon and gallium arsenide.

The class of materials are those disclosed by Smolenskii as having a layered perovskite-like structure, as discussed in the Background of the Invention. It has been realized that these materials are more than a substance which spontaneously forms in layers. This is seen most easily by an example. Strontium bismuth tantalate (SrBi2Ta2O9) can ...

... in the following definition: (B) a material having a localized structure, within a grain or other larger or smaller unit, which localized structure contains predominately repeatable units containing one or more perovskite-like layers and one or more intermediate non-perovskite-like layers spontaneously linked in an interdependent manner.

It is well-known that compounds having the perovskite structure may be described in terms of the general formula ABQ3, where A and B are cations and Q is an anion. In the ...

... flexible than the lattice of a ferroelectric material. Turning to FIG. 13, a layered superlattice material 92 is illustrated. Smolenskii recognized Pat. No. 5423285, \*

**FOCUS** 

that what we call the layered superlattice materials spontaneously form into layers 94 with a perovskite-like structure which alternate with layers 96 having a simpler structure. Depending on the material, the perovskite-like layers 94 may include one or a plurality of linked layers of perovskite-like octahedrons 90. As an example, FIG. 14 shows a unit cell of the material ABi2B2< + 5> 09, which is the formula for strontium bismuth tantalate (SrBi2Ta2O9) and other layered superlattice materials, such as tantalum, niobium, and tungsten, having a element with a valence of + 5 in the B-site. In this structure, each perovskite-like layer 94 includes two layers of octahedrons 90 which are separated by layers 96 of a material that does not have a perovskite-like structure. In this material the primitive unit cell consists of two perovskite layers 94 and two non-perovskite layers 96, since the structure shifts between the layers 98A and 98B. in FIG. ...

... 015, which is the formula for strontium bismuth titanate (SrBi4Ti4015) and other layered superlattice materials having an element, such as titanium, hafnium, and zirconium, having a valence of + 4 in the B-sites. In this material each the perovskite-like layer 94 has four layers of octahedrons 90.

It has been discovered that the layered superlattice materials catalogued by Smolenskii et al. are all likely candidates for fatigue free switching ferroelectrics and dielectric materials that are resistant to ...

... Sk xk < + sk> B1 y1 < + b1> B2 y2 < + b2> . . . B1 y1 < + b1> Q z < - 2> ,tm (1)

where Al, A2 . . . Aj represent A-site elements in the perovskite-like structure, which may be elements such as strontium, calcium, barium, bismuth, lead, and others S1, S2 . . . Sk represent superlattice generator elements, which usually is bismuth, but can also be materials such as yttrium, scandium, lanthanum, antimony, chromium, thallium, and other elements with a valence of + 3, B1, B2 . . . B1 represent B-site elements in the perovskite-like structure, which may be elements such as titanium, tantalum, hafnium, tungsten, niobium, zirconium, and other elements, and Q represents an anion, which generally is oxygen but may also be other elements, such as fluorine, ...

FOCUS - 34 OF 107 PATENTS

5,409,890

Apr. 25, 1995

Ξ,

The second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second secon

INVENTOR: Yamamoto, Susumu, Hyogo, Japan Kawabe, Nozomu, Hyogo, Japan Awazu, Tomoyuki, Hyogo, Japan Murai, Teruyuki, Hyogo, Japan

... term quasi-perovskite type means a structure which can be considered to have such a crystal structure that is similar to perovskite-type oxides and includes an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

The sintering operation of the powder mixture is carried out at temperature which is higher than 6000 C. but is not higher than the lowest melting point of any component in the material powder to be sintered. If the sintering temperature exceeds the ...

FOCUS - 35 OF 107 PATENTS

5,401,715

<=2> GET 1st DRAWING SHEET OF 1

Mar. 28, 1995

Semiconductor substrate having a superconducting thin film

INVENTOR: Itozaki, Hideo, Hyogo, Japan Harada, Keizo, Hyogo, Japan Fujimori, Naoji, Hyogo, Japan Yazu, Shuji, Hyogo, Japan Jodai, Tetsuji, Hyogo, Japan

#### DETDESC:

... term quasi-perovskite type means a structure which can be considered to have such a crystal structure that is similar to perovskite-type oxides and includes an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

An atomic ratio of the lanthanide element "Ln": Ba: Cu is preferably 1:2:3 as is defined by the formula but the atomic ratio is not restricted strictly to this ratio. In fact, the other compound oxides having ... FOCUS - 36 OF 107 PATENTS

5,389,603

<=2> GET 1st DRAWING SHEET OF 5

Feb. 14, 1995

Oxide superconductors, and devices and systems comprising such a superconductor

INVENTOR: Batlogg, Bertram J., New Providence, New Jersey Cava, Robert J., Bridgewater, New Jersey

... microscopy indicate a basically orthorhombic crystal structure, but there are also indications that, at least for some of the inventive compounds, the structure may be weakly monoclinic. Both of these possibilities are intended to be included in the term "perovskite-like" or analogous terms. Diffraction studies have also revealed the presence of a variety of long period long range and the second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second second s ordered superlattices (typically in the ab plane).

ماند و الداريج الرافيدة عل<del>اه و و</del>يد الرابع وفريون بأجزا ب<u>ن ميني</u>ين من أثر الإربيسمسور بعدر الإرار

FIG. 2 shows the field (225 Oe)-cooled ...

We claim:

- [\*1] 1. An article comprising a superconductive element comprising at least one superconductive material having a perovskite-like crystal structure and nominal formula (Pb2A2 Cu')BCu2O8 + delta with (A selected from the group consisting of Sr, Ba, Sr and Ba, Sr and Ca, and Sr, Ba and Ca; Cu' is selected from the group consisting of ...
- ... [\*1] parallel to the ab- plane; and wherein the composition is selected such that the superconductive material has a transition temperature of at least about 30K.
- [ $^*2$ ] 2. An article comprising a superconductive element comprising at least one superconductive material having a perovskite-like crystal structure and nominal formula (X2A2Cu') BCu2O8 + delta , where X is selected from the group consisting of Pb, Pb and Bi, Pb and Tl, and Pb, Bi and Tl, with X being at least 50 atomic % of ...

FOCUS - 37 OF 107 PATENTS

5,362,710

<=2> GET 1st DRAWING SHEET OF 2

Nov. 8, 1994

Process for preparing high Tc superconducting material

INVENTOR: Fujita, Nobuhiko, Hyogo, Japan Kobayashi, Tadakazu, Hyogo, Japan Itozaki, Hideo, Hyogo, Japan Tanaka, Saburo, Hyogo, Japan Yazu, Shuji, Hyogo, Japan Jodai, Tetsuji, Hyogo, Japan

#### STIM .

... quasi-perovskite type oxide means a structure which can be considered to have such a crystal structure that is similar to perovskite-type oxides and includes an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

The present invention also provides a process for producing the abovementioned superconducting material, characterized by sintering a mixture of the following powders:

an oxide, carbonate, nitrate or sulfate of one element "A" selected from ... FOCUS - 38 OF 107 PATENTS

5,356,674

<=2> GET 1st DRAWING SHEET OF 2

Oct. 18, 1994

Process for applying ceramic coatings using a plasma jet carrying a free form non-metallic element

INVENTOR: Henne, Rudolf, Boeblingen, Federal Republic of Germany Weber, Winfried, Leinfelden-Echterdingen, Federal Republic of Germany Schiller, Guenter, Gerlingen, Federal Republic of Germany Schnurnberger, Werner, Stuttgart, Federal Republic of Germany Kabs, Michael, Hanau, Federal Republic of Germany

### SUM:

... materials are oxidized materials, for example, spinels and perovskites on a nickel or cobalt or nickel-cobalt basis. It is, however, also conceivable to apply all possible kinds of spinels and perovskites in accordance with the inventive process. This also applies to spinel-like and perovskite-like compounds and to non oxidized compounds, for example, nitrides, halides, carbides, etc., with nitrogen or halogens or also non-metallic compounds,

methane or acetylene then being carried along as non-metallic element by the ... FOCUS - 39 OF 107 PATENTS

5,354,733

<=2> GET 1st DRAWING SHEET OF 21

Oct. 11, 1994

Copper oxide superconductor containing carbonate radicals

INVENTOR: Kinoshita, Kyoichi, Hoya, Japan Yamada, Tomoaki, Higashimurayama, Japan

SUM:

... 2. Description of the Prior Art

Several types of copper oxide superconductors have been discovered since high-T c superconductivity was detected in the La-Ba-Cu-O system. Superconductivity would arise from the layered perovskite-like structure having CuO6 octahedra, or CuO5 pyramids, or CuO2 square planes as a building unit. The layered perovskite-like structure and a sufficient carrier concentration of the material are essential factors for making the material superconducting as indicated by Osamura & Zhang (Japan.J.Appl.Phys.26, L2094-L2096, 1987). ... FOCUS - 40 OF 107 PATENTS

5,340,796

<=2> GET 1st DRAWING SHEET OF 5

Aug. 23, 1994

Oxide superconductor comprising Cu, Bi, Ca and Sr

INVENTOR: Cava, Robert J., Bridgewater, New Jersey Sunshine, Steven A., Berkeley Heights, New Jersey

ABST:

Novel superconductive oxides are disclosed. The oxides all have layered perovskite-like crystal structure and manifest superconductivity above about 77K. An exemplary material has composition Bi2.2Sr2Ca0.8Cu2O8. Other materials are described by the nominal formula X 2 + x M n - x Cu n - ...

SUM:

... high temperature superconductors has been reported since publication of the above seminal papers. Most of the work deals with YBa2Cu3O x (the so-called 1-2-3 compound) and related compounds.

In all of these compounds the superconducting phase is perovskite-like, typically having orthorhombic crystal structure, and the compounds that exhibit high (i.e., T c > 77K) temperature superconductivity generally contain one or more rare earth elements.

The discovery of high T c superconductivity in some ...

... likely to be stable high T c superconductors, with T c s likely to be above 100K.

The novel phases all have a crystal structure that is closely related to that of the above described 80K compound and thus are perovskite-like. They differ from each other essentially only in the number of crystal planes between the two Bi-O double planes that bound the unit cell in the c-direction, or by the size of the supercell. The composition of the ...

... in added layers of M and Cu between the Bi-O double layers and are expected to result in one or more phases of stable high T c superconductive

material. All of the inventive phases have layered perovskite-like crystal structure. and the existence of relatively weak bonding between at least some layers may be the cause of the observed relatively high ductility of the inventive materials. It will be appreciated that by "perovskite-like" we mean not only the prototypical, truly cubic structure, but very significantly distortions therefrom. Material specification in accordance with the invention depends upon the nature of the intended use. For power transmission, or any other currentcarrying Pat. No. 5340796, \* What is claimed is: 1. An article comprising material perovskite-like structure and of [\*1] nominal composition X 2 + x M 4 - x Cu3010 + 0.5 +/- delta , where [x = p/q <0.4, and p and q are positive integers] 0 < = x < 0.4, X is Bi and Pb, ... FOCUS - 41 OF 107 PATENTS 5,338,721 <=2> GET 1st DRAWING SHEET OF 5 Aug. 16, 1994 Process for manufacturing a superconducting composite

PAGE

**FOCUS** 

INVENTOR: Yamamoto, Susumu, Hyogo, Japan Murai, Teruyuki, Hyogo, Japan Kawabe, Nozomu, Hyogo, Japan Awazu, Tomoyuki, Hyogo, Japan Yazu, Shuji, Hyogo, Japan Jodai, Tetsuji, Hyogo, Japan

### DETDESC:

... quasi-perovskite type structure" means any oxide that can be considered to have such a crystal structure that is similar to perovskite-type oxides and may include an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

In practice, the element alpha is preferably selected from Ba, Sr and/or Ca and the element beta is preferably selected from Y, La and/or lanthanid such as Sc, Ce, Gd, Ho, Er, Tm, Yb, Lu and the ... FOCUS - 42 OF 107 PATENTS

5,332,722

<=2> GET 1st DRAWING SHEET OF 3

Jul. 26, 1994

Nonvolatile memory element composed of combined superconductor ring and MOSFET

INVENTOR: Fujihira, Mitsuka, Yokohama, Japan

... term quasi-perovskite type means a structure which can be considered to have such a crystal structure that is similar to perovskite-type oxides and includes an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

41-11-54 - <del>1-</del> Another superconducting compound oxide which can be used by the present invention is represented by the general formula:

(M,Sr)2CuO 4- delta in which M stands for Y or La and ...

FOCUS - 43 OF 107 PATENTS

5,328,892

Jul. 12, 1994

Oxide superconductor composition and a process for the production thereof

INVENTOR: Manako, Takashi, Tokyo, Japan Shimakawa, Yuichi, Tokyo, Japan Kubo, Yoshimi, Tokyo, Japan

SUM:

... following formulae:

T1Sr 3 - x Y x Cu207(IA)

wherein 0.1 </= x </= 1, and

T1Sr 4 -  $\times$  Y  $\times$  Cu309(IB)

wherein 0.1 < = x < = 2. Unit cells of the layered perovskite-like crystal structures of these compositions of the formulae (IA) and (IB) may be shown respectively as follows:

T10/Sr0/Cu02/Sr or Y/Cu02/Sr0(IX)

T10/Sr0/Cu02/Sr or Y/ ..

FOCUS - 44 OF 107 PATENTS

5,296,458

<=2> GET 1st DRAWING SHEET OF 4

Mar. 22, 1994

Epitaxy of high T c superconducting films on (001) silicon surface

INVENTOR: Himpsel, Franz J., Mt. Kisco, New York

SUM:

... first showed superconducting behavior in mixed copper-oxides, typically including rare earth and/or rare earth-like elements and alkaline earth elements, for example La, Ba, Sr, . . , and having a perovskite-like structure. Materials including the so called "1-2-3" phase in the Y-Ba-Cu-O system have been found to exhibit a superconducting transition temperature in excess of 77K.

R. B. ...

FOCUS - 45 OF 107 PATENTS

5,286,712

<=2> GET 1st DRAWING SHEET OF 2

Feb. 15, 1994

High TC superconducting film

INVENTOR: Fujita, Nobuhiko, Hyogo, Japan Kobayashi, Tadakazu, Hyogo, Japan

Itozaki, Hideo, Hyogo, Japan Tanaka, Saburo, Hyogo, Japan Yazu, Shuji, Hyogo, Japan Jodai, Tetsuji, Hyogo, Japan

#### SUM:

... quasi-perovskite type oxide means a structure which can be considered to have such a crystal structure that is similar to perovskite-type oxides and includes an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

The present invention also provides a process for producing the abovementioned superconducting material, characterized by sintering a mixture of the following powders:

an oxide, carbonate, nitrate or sulfate of one element "A" selected from ...
FOCUS - 46 OF 107 PATENTS

5,283,465

<=2> GET 1st DRAWING SHEET OF 5

Feb. 1, 1994

Superconducting lead on integrated circuit

INVENTOR: Yamazaki, Shunpei, Tokyo, Japan

### **DETDESC:**

... subjected to supplemental annealing at 5000-6000 C. for 1-2 hours as illustrated in FIG. 1(B). The supplemental annealing allows the superconducting ceramic material to form a modulated perovskite-like structure and, as a result, a high critical temperature is realized. On the substrate, there are provided superconducting leads 10 and 10' for interconnection among devices and contacts formed in or on the semiconductor substrate and a ...

FOCUS - 47 OF 107 PATENTS

5,278,140

<=2> GET 1st DRAWING SHEET OF 5

Jan. 11, 1994

Method for forming grain boundary junction devices using high T c superconductors

INVENTOR: Chaudhari, Praveen, Briarcliff Manor, New York
Chi, Cheng-Chung J., Yorktown Heights, New York
Dimos, Duane B., Montclair, New Jersey
Mannhart, Jochen D., Metzingen, New York, Federal Republic of Germany
Tsuei, Chang C., Chappaqua, New York

#### SUM:

... first showed superconducting behavior in mixed copper-oxides, typically including rare earth and/or rare earth-like elements and alkaline earth elements, for example La, Ba, Sr, . . . , and having a perovskite-like structure. Materials including the so called "1-2-3" phase in the Y-Ba-Cu-O system have been found to exhibit a superconducting transition temperature in excess of 77K. R. B. ...

FOCUS - 48 OF 107 PATENTS

5,252,547

<=2> GET 1st DRAWING SHEET OF 1

Oct. 12, 1993

Method of forming an inorganic protective layer on an oxide superconducting film

INVENTOR: Itozaki, Hideo, Hyogo, Japan Tanaka, Saburo, Hyogo, Japan Fujita, Nobuhiko, Hyogo, Japan Yazu, Shuji, Hyogo, Japan Jodai, Tetsuji, Hyogo, Japan

#### SUM:

... term of quasi-perovskite type means a structure which can be considered to have such a crystal structure that is similar to Perovskite-type oxides and includes an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

The superconducting thin film may be also another type of superconductor consisting mainly of a compound oxide represented by the formula:

THETA 4( PHI 1 - q ,Ca q ) m Cu ... FOCUS - 49 OF 107 PATENTS

5,249,525

<=2> GET 1st DRAWING SHEET OF 11

Oct. 5, 1993

Spark-discharge lithography plates containing image-support pigments

INVENTOR: Lewis, Thomas E., E. Hampstead, New Hampshire Nowak, Michael T., Gardner, Massachusetts

#### DETDESC:

... A perspective view of the first layer, labeled "Layer 0", appears in FIG. 6E. As shown in these figures, the spinel structure contains a number of octahedral sites for metal ions. Like perovskite structures spinels may also be defective, an example being gamma-Fe203. A spinel structure may also be intergrown with other structures.

In spinel compounds useful as image-support pigments, the ... FOCUS - 50 OF 107 PATENTS

5,244,874

Sep. 14, 1993

Process for producing an elongated superconductor

INVENTOR: Yamamoto, Susumu, Hyogo, Japan Kawabe, Nozomu, Hyogo, Japan Awazu, Tomoyuki, Hyogo, Japan

## DETDESC:

... term quasi-perovskite type means a structure which can be considered to have such a crystal structure that is similar to perovskite-type oxides and includes an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

Another superconducting compound oxide which can be prepared by the present invention is represented by the general formula:

(M, Sr)2CuO 4 - delta

FOCUS - 51 OF 107 PATENTS

5,241,191

## <=2> GET 1st DRAWING SHEET OF 1

Aug. 31, 1993

Cubic perovskite crystal structure, a process of preparing the crystal structure, and articles constructed from the crystal structure

INVENTOR: Agostinelli, John A., Rochester, New York Chen, Samuel, Penfield, New York

## DETDESC:

... 1, PA-2, PA-3, PA-4 and PA-5, cited above and here incorporated by reference, can be employed. Highly compatible substrates are materials that themselves exhibit a perovskite or perovskite-like crystal structure. Strontium titanate is an example of a perovskite crystal structure which is specifically preferred for use as a substrate. Lanthanum aluminate (LaAlO3), lanthanum gallium oxide (LaGaO3) and potassium tantalate are ...

FOCUS - 52 OF 107 PATENTS

5,236,894

Aug. 17, 1993

Process for producing a superconducting thin film at relatively low temperature

INVENTOR: Tanaka, Saburo, Itami, Japan Itozaki, Hideo, Itami, Japan Higaki, Kenjiro, Itami, Japan Yazu, Shuji, Itami, Japan Jodai, Tetsuji, Itami, Japan

### SUM:

... crystal structure. The term quasi-perovskite type means a structure which can be considered to be similar to perovskite-type oxides and includes an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

Still another example of the above-mentioned compound oxide is compound oxides represented by the general formula:

THETA 4( PHI 1 - q , Ca q ) m Cu n 0 p + ... FOCUS - 53 OF 107 PATENTS

5,221,660

<=2> GET 1st DRAWING SHEET OF 1

Jun. 22, 1993

Semiconductor substrate having a superconducting thin film

INVENTOR: Itozaki, Hideo, Hyogo, Japan Harada, Keizo, Hyogo, Japan Fujimori, Naoji, Hyogo, Japan Yazu, Shuji, Hyogo, Japan Jodai, Tetsuji, Hyogo, Japan

## DETDESC:

... term quasi-perovskite type means a structure which can be considered to have such a crystal structure that is similar to perovskite-type oxides and includes an orthorhombically distorted perovskite or a distorted oxygen-deficient perovskite or the like.

An atomic ratio of the lanthanide element "Ln":Ba:Cu is preferably 1:2:3 as is defined by the formula but the atomic ratio is not restricted strictly to this ratio. In fact, the other compound oxides having ...

FOCUS - 54 OF 107 PATENTS

5,212,148

<=2> GET 1st DRAWING SHEET OF 1

May 18, 1993

Method for manufacturing oxide superconducting films by laser evaporation

INVENTOR: Roas, Bernhard, Erlangen, Federal Republic of Germany Endres, Gerhard, Forchheim, Federal Republic of Germany Schultz, Ludwig, Bubenreuth, Federal Republic of Germany

SUM:

... yet exactly established. This initial product is then converted, by applying a heat and oxygen treatment, into the material with the desired superconducting phase.

The superconductive metal-oxide phases, to be obtained in this manner, can have perovskite-like crystal structures and, in the case of YBa2Cu3O 7 - x, whereby 0 < x < 0.5, have an orthorhomic structure (compare, for example, "Europhysics Letters", Vol. 3, No. 12, Jun. 15, 1987, pages ... FOCUS - 55 OF 107 PATENTS

5,183,799

<=2> GET 1st DRAWING SHEET OF 16

Feb. 2, 1993

Superconducting materials including La-Sr-Nb-0, Y-Ba-Nb-0, La-Sr-Nb-Cu-0, and Y-Ba-Nb-Cu-0

INVENTOR: Ogushi, Tetsuya, Kagoshima, Japan Hakuraku, Yoshinori, Kagoshima, Japan Ogata, Hisanao, Ibraki, Japan

ABST:

... V, Nb, Ta, T, Zr or Hf; 0 < x < 1; 0 < z < 1; i = 1, 3/2 or 2; 0 < y </=4; Gis F, Cl or N; delta is oxygen defect, and having a perovskite-like crystal structure, show superconductivity at a temperature higher than the liquid nitrogen temperature.

SUM:

BACKGROUND OF THE INVENTION

This invention relates to a superconducting material having a perovskite-like crystal structure and a superconducting part using the same, particularly to a superconducting material suitable for having a high superconducting transition temperature (Tc), and a process for producing the same.

Heretofore, ...

DETDESC:

DESCRIPTION OF THE PREFERRED EMBODIMENTS

The superconducting materials of this invention have a perovskite-like crystal structure and represented by the formulae:

The Bupercom

```
(L x A 1 - x ) i MO y( 1)
(L x A 1 - x ) i M 1 - z Cu z ...
```

... by laminating this superconducting material with other films of electrical insulating material. It is preferable to laminate a plurality of film-like layers alternately, respectively. Further, it is preferable to use as an insulating material a perovskite-like ceramic of the same series.

Further, in the above-mentioned formulae (1) and (2), a total of valence number (p) of L, A and M, or L, A, M and Cu, and the valence number y of ...

... OMITTED p SYMBOL OMITTED = SYMBOL OMITTED y SYMBOL OMITTED +/- 0.5
Pat. No. 5183799, \*

**FOCUS** 

Further, it is preferable to include M of the valence of two.

More in detail, the material represented by the formula (1) has a perovskite-like crystal structure and has as the L element at least one element selected from the group consisting of scandium (Sc), yitrium (Y), and lanthanide elements of atomic numbers 57 to 71 (La to Lu) belonging to the group ...

... Ta) belonging to the group Vb of the periodic table and titanium (Ti), zirconium (Zr) and hafnium (Hf) belonging to the group IVb of the periodic table, these element being able to include Cu.

The oxide superconducting material having the perovskite-like crystal structure of this invention has as a fundamental constitution an octahedron having the M element which is an atom belonging to the group Vb or IVb as its center and 6 oxygen atoms. Since this material has defect of oxygen, that is, one or ...

... a mutual action of strong attraction necessary for forming a hole pair or electron pair showing a superconducting phenomenon at a temperature of 150K or higher.

The oxide superconducting material of this invention has the perovskite-like crystal structure as shown in FIGS. 1 and 2. These drawings show unit lattices of the materials represented by the formulae:

```
(L x A 1 - x ) i MO y( 1) and ( ...
```

... formula (4) with at least one element selected from those of the group IVb and Vb, the total amount of the elements of the group IVb and Vb can exceed the amount of Cu.

It is also possible to produce an oxide superconducting powder having a perovskite-like crystal structure containing M element mainly by mixing a powder of oxide material represented by (L x A 1 - x ) i CuO y , wherein x is 0 < x < 1; ...

... Cu:M = 1:1, carrying out substitution reaction between Cu and M element in vacuum, and finally pulverizing the final reaction product.

It is further possible to produce an oxide superconducting powder having a perovskite-like crystal structure and containing M element mainly by depositing in vacuum a film of pure metal of M element selected from the elements of groups IVb and Vb on outer surface of oxide ceramic ...

... 1, 3/2 or 2; y is 0 < y </= 4, containing the M element mainly (M being



# Attachment D

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Jul. 28, 1992

Solid compositions for fuel cells, sensors and catalysts

INVENTOR: Madou, Marc J., Palo Alto, California Otagawa, Takaaki, Fremont, California Sher, Arden, Foster City, California

... [\*12] selected from lanthanum, cerium, neodymium, praseodymium, or scandium, B is independently selected from strontium, calcium, barium or magnesium, Q is independently selected from nickel, cobalt, iron or manganese, and y is between about 0.0001 and 1, wherein the perovskite or perovskite-type structure has an average size and distribution of between about 50 and 200 Angstroms in diameter; and the composite layer of between about 25 and 1000 microns in thickness;

**PAGE** 

LEVEL 1 - 2 OF 2 PATENTS

4,948,680

<=2> GET 1st DRAWING SHEET OF 26

Aug. 14, 1990

Solid compositions for fuel cell electrolytes

INVENTOR: Madou, Marc J., Palo Alto, California Otagawa, Takaaki, Fremont, California Sher, Arden, Foster City, California

... [\*25] 1.5 and d is between 0.001 and less than or equal to 3,

wherein either the first electrode material (C) or second electrode material (A') comprises

A 1 - x B x Q0 3

having a perovskite or perovskite-type structure as an electrode catalyst in combination with

A 1 - x B x Z

as a polycrystalline solid electrolyte wherein

A is independently selected from lanthanum, cerium, neodymium, praseodymium or scandium,

\* 2 PAGES 36 LINES JOB 53252 100G6J \* 12:53 P.M. STARTED 12:53 P.M. ENDED 11/24/97 \*

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# $\begin{array}{c} Perovskites \ and \ High \ T_c \\ Superconductors \end{array}$

bу

Francis S. Galasso

United Technologies Research Laboratories East Hartford, Connecticut USA

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these compounds as ribed. In addition, a les oxides with the

tic bubble research, high dielectric conic memory work and en conducted for the of interest in devices to been investigated

te more interest in in that the greatest the discovery of the ik is devoted to this

#### Chapter II

# Structure of Perovskite-type Compounds

Most of the compounds with the general formula ABO<sub>3</sub> have the perovskite structure. The atomic arrangement in this structure was first found for the mineral perovskite, CaTiO<sub>3</sub>. It was thought that the unit cell of CaTiO<sub>3</sub> could be represented by calcium ions at the corners of a cube with titanium ions at the body center and oxygen ions at the center of the faces (Fig. 2.1). This simple cubic structure has retained the name perovskite, even though CaTiO<sub>3</sub> was later determined to be orthorhombic by Megaw.<sup>(1)</sup> Through the years it has been found that very few perovskite-type oxides have the simple cubic structure at room temperature, but many assume this ideal structure at higher temperatures.

In the perovskite structure, the A cation is coordinated with twelve oxygen ions and the B cation with six. Thus, the A cation is normally found to be somewhat larger than the B cation. In order to have contact between the A, B, and O ions,  $R_A + R_O$  should equal  $\sqrt{2(R_B + R_O)}$ , where  $R_A$ ,  $R_B$  and  $R_O$  are the ionic radii. Goldschmidt<sup>(2)</sup> has shown that the cubic perovskite structure is stable only if a tolerance factor, t defined by  $R_A + R_O = t\sqrt{2(R_B + R_O)}$ , has an approximate range of 0.8 < t < 0.9, and a somewhat larger range for distorted perovskite structures. It should be noted that conflicting reports in the literature make it difficult to assign the correct unit cell dimensions for these distorted perovskite structures.

The ternary perovskite-type oxides described in this chapter will be divided into A<sup>1+</sup>B<sup>5+</sup>O<sub>3</sub>, A<sup>2+</sup>B<sup>4+</sup>O<sub>3</sub>, A<sup>3+</sup>B<sup>3+</sup>O<sub>3</sub> types and oxygen- and cation-deficient phases. The oxygen- and cation-deficient phases will be regarded as those which contain considerable vacancies and not those phases which are only slightly non-stoichiometric. Many of these contain B ions of one element in two valence states and should

Attachment

# COPPER OXIDE SUPERCONDUCTORS

Charles P. Poole, Jr. Timir Datta
Horacio A. Farach

with help from

M. M. Rigney C. R. Sanders

Department of Physics and Astronomy University of South Carolina Columbia, South Carolina



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#### 78 CRYSTALLOGRAPHIC STRUCTURES

tetragonal phase, and the metal-to-insulator transition occurs at the tetragonal-to-orthorhombic phase boundary  $x \approx 0.35$  (Matt7, Slei1).

#### D. PEROVSKITE-TYPE SUPERCONDUCTING STRUCTURES

In their first report on high-temperature superconductors Bednorz and Müller referred to their samples as "metallic, oxygen deficient . . . perovskite like mixed valent copper compounds." Subsequent work has confirmed that the new superconductors do indeed have these characteristics. In this section we will comment on their perovskite-like aspects.

#### 1. Atom Sizes

In the oxide superconductors Cu replaces the Ti<sup>4+</sup> ions (0.68 Å) of perovskite, and in most cases retains the CuO<sub>2</sub> layering with two oxygens per copper in the layer. Other cationic replacements tend to be Bi, Ca, La, Sr, Tl, and Y for the larger Ba, forming "layers" containing only one oxygen or none per cation. We see from the following list of ionic radii

that there are four size groups, with all other cations significantly smaller then the Ba of perovskite. The common feature of  $CuO_2$  layers that are planar or close to planar establishes a fairly uniform lattice size in the a,b plane. The parameters of the compounds LaSrCuO (a=b=3.77 Å), YBaCuO (a=3.83 Å, b=3.89 Å), BiSrCaCuO (a=b=3.82 Å), and TlBaCaCuO (a=b=3.86 Å) are all between the ideal fcc oxygen lattice value of 3.73 Å and the perovskite one of 4.01 Å.

Table VI-2 gives the ionic radii of the positively charged ions of various elements of the periodic table. These radii are useful for estimating changes in lattice constant when ionic substitutions are made in existing structures. They also provide some insight into which types of substitutions will be most favorable.

ion occurs at the tetragonal-Slei1).

#### **STRUCTURES**

uctors Bednorz and Müller icient . . . perovskite like has confirmed that the new cs. In this section we will

ns (0.68 Å) of perovskite, oxygens per copper in the La, Sr, Tl, and Y for the en or none per cation. We

(VI-4)

ignificantly smaller then s that are planar or close a,b plane. The parameaCuO (a = 3.83 Å, b =CuO (a = b = 3.86 Å) 3 Å and the perovskite

ged ions of various eleimating changes in latig structures. They also ill be most favorable.

TABLE VI-2. Ionic Radii in Angstroms of Selected Elements for Various Positive Charge States"

	Flores				1.4		
<u>z</u>	Element	+1	+2	+3	+4	+5	+6
			Alk	ali			
3	Li	0.68					
11	Na	0.97					
19	K	1.33					
37	Rb	1.47					
55	Cs	1.67					
			Alkaline	eurths			
4	Be	0.44	0.35		•		
12	Mg	0.82	0.66				
20	Ca	1.18	0.99				
38	Sr		1.12				
56	Ва	1.53	1.34				
			Group	111			
5	В	0.35		0.23			*
13	Αl			0.51			
31	Ga	0.81		0.62			
49	In			0.81			
81	TI	1.47		0.95			
			Group	IV			
6	C				0.16		
14	Si	0.65			0.42		
32	Ge		0.73		0.53		
50	Sn		0.93		0.71		
82	Pb		1.20		0.84		
			Group	, <i>v</i>			
15	P			0.44		0.35	
33	As			0.58		0.46	
51	Sb	0.89		0.76		0.62	
83	Bi	0.98		0.96		0.74	
		~	Chalcogo	nides			
16	S				0.37		0.30
34	Se	0.66			0.50		0.42
52	Te	0.82			0.70		0.56
		Firs	st transition	series (3d°	,		
21	Sc			0.81			
22	Ti	0.96	0.94	0.76	0.68		
23	· V		0.88	0.74	0.63	0.59	
24	Cr	0.81	0.89	0.63			0.52
25	Мп		0.80	0.66	0.60		

+(	+5	<del>+</del> 4	+3	+2	+1	Element	<u>z</u>
			0.64	0.74		Fe	26
			0.63	0.72		Co	27
				0.69		Ni	28
				0.72	0.96	Cu	29
				0.74	0.88	Zn	30
		<b>"</b> /	n series (4d'	nd transitio	Seco		
			0.94			Y	39
		0.79			1.09	Zr	40
	0.69	0.74			1.00	Nb	41
0.6		0.70			0.93	Mo	42
		-				Tc	43
		0.67				Ru	44
			0.68			Rh	45
		0.65		0.80		Pd	46
				0.89	1.26	Ag	47
				0.97	1.14	Cd	48
			series (5d°)	d transition	Thir		
		0.78				Hf	72
	0.68					Ta	73
0.62	0.00	0.70				W	74
0.02		0.72				Re	75
0.69		0.88				Os	76
0.0		0.68				lr	77
		0.65		0.80		Pt	78
		0.00	0.85		1.37	Au	79
				1.10	1.27	Hg	80
			(4f")	Rare earths			
			1.14		1.39	La	57
		0.94	1.07		1.27	Ce	58
		0.92	1.06			Pr	59
			1.04			Nd	50
			1.06			Pm	1
			1.00			Sm	52
			0.98			Eu	3
			0.62			Gd 🔪	·4
		0.81	0.93			Tb	5
			0.92			Dy	6
			0.91			Ho	7
			0.89			Er	8
			0.87			Tm	9
			0.86			Yb	0
			0.85			Lu	1

<sup>\*</sup>Three anion radii are 1.32 for  $O^{2-}$ , 1.33 for  $F^-$ , and 1.84 for  $S^{2-}$  (Handbook of Chemistry and Physics).

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Attachment I

# COPPER OXIDE SUPERCONDUCTORS

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with help from

M. M. Rigney C. R. Sanders

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## VI

### **CRYSTALLOGRAPHIC STRUCTURES**

#### A. INTRODUCTION

To properly understand the mechanisms that bring about the superconducting state in particular materials it is necessary to know the structures of the compounds that exhibit this phenomenon. Single-crystal structure studies have been carried out to determine the dimensions of the unit cell, the locations of the atoms in this cell, electronic charge distributions, and the possible presence of atomic irregularities. Neutron powder diffraction has also provided much of the detailed structure information found in this chapter (e.g., Antso, Beech, Cappo, Coxzz, Davil, Dayzz, Greed, John 4. Jorge, Jorgl, Paulz, Torar, Vakni, Yamag, Yanz 2). More routine X-ray powder pattern measurements which can identify a known structure and provide the unit cell dimensions are useful for checking the quality of samples, as was explained in Section V-1.

The numerical values of quantities such as lattice parameters and bond lengths show some variation in the literature, and many of our quoted values will be typical ones. Much of the quantitative structural information is organized in the tables.

In the beginning of this chapter we will introduce the perovskite structure and indicate how it is related to the oxide superconductors. Then we will describe the 21 structure of LaSrCuO and the 123 structure of YBaCuO, we will show how each is generated from a perovskite prototype, and we will clarify its layering scheme. The chapter will end with descriptions of the structures of the newer high-transition-temperature bismuth and thallium compounds.

#### **B. PEROVSKITES**

Much has been written about the oxide superconductor compounds being perovskite types, so we will begin with a description of the perovskite structure. This will permit us to develop some of the notation to be used in describing the structures of the superconductors themselves.

#### 1. Cubic Form

Above 200°C barium titanate crystallizes in the perovskite structure, which is cubic, so the three lattice parameters are all equal (i.e., a = b = c). The unit cell contains one formula unit BaTiO<sub>3</sub> and the atoms are located in the following special positions (Wyck2, p. 390):

Ba 
$$(1a)$$
  $\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$   
Ti  $(1b)$   $0,0,0$   
O  $(3c)$   $0,0,\frac{1}{2}; 0,\frac{1}{2},0; \frac{1}{2},0,0$  (VI-1)

where we have employed the crystallographic notation (1a) for an a-type lattice site which contains one atom, (3c) for a c-type lattice site which contains three atoms, and so on. Each atomic position is given by three coordinates, such as  $0,0,\frac{1}{2}$  for the oxygen located at x=0,y=0,z=0.5a. This arrangement corresponds to placing a titanium atom on each apex. a barium atom in the body center, and an oxygen atom on the center of each edge of the cube, as illustrated on Fig. VI-1. We see from the figure that the barium atoms are 12-fold coordinated and the titaniums have sixfold (octahedral) coordination. The lattice constant or length of the unit cell is a=4.0118 Å at 201°C. The crystallographic space group is Pm3m,  $O_1^1$ .

An alternate way to represent this structure, which is commonly used in solidstate texts and in crystallography monographs (e.g., Wyck2), is to locate the

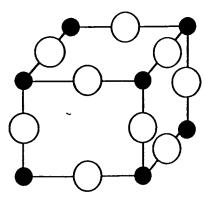


Fig. VI-1. Perovskite cubic unit cell showing titanium on the apices and oxygen in the edge-centered positions. Barium, which is in the body center, is not shown.

**CTURES** 

the superconducting uctures of the comre studies have been the locations of the possible presence of rovided much of the 1tso, Beech, Cappo, rar, Vakni, Yamag, which can identify a ful for checking the

ameters and bond r quoted values will tion is organized in

skite structure and we will describe the we will show how clarify its layering tures of the newer ds. origin at the barium site; this places titanium in the center and the oxygens on the centers of the cube faces. The representation (Eq. VI-1) given above is more convenient for comparison with the structures of the oxide superconductors.

The compound LaBaCu<sub>2</sub>O<sub>5</sub> was found to have a cubic perovskite subcell with the lattice parameter a = 3.917 Å (Sishe).

#### 2. Tetragonal Form

At room temperature barium titanate is tetragonal with the unit cell dimensions a=3.9947 Å and c=4.0336 Å, which is close to cubic. For this lower symmetry the oxygens are assigned to two different sites, a single site along the side edges and a twofold one at the top and bottom. The atomic positions (Wyck2, p. 401)

Ba 
$$\frac{1}{2}, \frac{1}{2}, 0.488$$
  
Ti 0,0,0  
O(1) 0,0,0.511 (VI-2)  
O(2)  $0, \frac{1}{2}, -0.026; \frac{1}{2}, 0, -0.026$ 

are shown in Fig. VI-2. The distortions from the ideal structure of Fig. VI-1 are exaggerated on this sketch. We will see later that a similar distortion occurs in the YBaCuO structure. The cubic and tetragonal atom arrangements (VI-1) and (VI-2) are compared in Table VI-1, and we see from this table that the deviation from cubic symmetry is actually quite small.

#### 3. Orthorhombic Form

When barium titanate is cooled below 5°C it undergoes a transition with a further lowering of the symmetry to the orthorhombic space group Amm2,  $C_{2v}$ , and

TABLE VI-1. Comparison of Atom Positions of BaTiO<sub>3</sub> in Its Cubic, Tetragonal and Orthorhombic Forms"

_		Cubic and	Tetragonal	Cubic	Tetragonal	Orthorhombic
Group	Atom	x	y	z	z	z
	( Ti	0	0	1	1	1
TiO <sub>2</sub>	<b>}</b> O	0	1/2	1	0.974	i
(	Ço	1/2 ~	Ō	1	0.974	i
BaO	{ O	0	0	1/2	0.511	į.
2	( Ba	1/2	1/2	1 7	0.488	2 1
	(Ti	0	0	ō	0	0
TiO <sub>2</sub>	} O	0	1/2	0	-0.026	Ö
	O	1/2	0	0	-0.026	Ŏ

<sup>&</sup>quot;The x and y coordinates are the same for both positions. The orthorhombic form z coordinates are also given (Wyck2, pp. 390, 401, 405).

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(VI-2)

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transition with a furoup Amm2, C<sub>2v</sub>, and

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ıl	Orthorhombic
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	0

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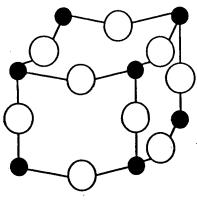


Fig. VI-2. Perovskite tetragonal unit cell showing the puckering of the Ti-O layers.

an enlargement of the unit cell to accommodate two formula units  $(BaTiO_3)_2$ . The enlarged cell is rotated by 45° relative to the higher-temperature ones, as shown on Fig. VI-3, and therefore its a and b lattice parameters are larger by the factor  $\sqrt{2}$ . The three lattice constants are  $a = 5.669 = 4.009\sqrt{2}$  Å,  $b = 5.682 = 4.018\sqrt{2}$  Å, and c = 3.990 Å. There are no longer any special sites, and the atomic positions are (Wyck2, p. 405):

Ba (2a) 
$$0,\frac{1}{2},\frac{1}{2}; \frac{1}{2},0,\frac{1}{2}$$
  
Ti (2b)  $0,u+\frac{1}{2},0; \frac{1}{2},u,0$  with  $u=0.510$   
O(1) (2a)  $0,u+\frac{1}{2},\frac{1}{2}; \frac{1}{2},u,\frac{1}{2}$  with  $u=0.490$   
O(2) (4e)  $u,v+\frac{1}{2},0; -u,v+\frac{1}{2},0; u+\frac{1}{2},v,0; -u+\frac{1}{2},v,0$   
with  $u=0.253, v=0.237$ 

where u = 0 for Ba.

One should note that in Eq. (VI-3) Ba and O(1) are in the same (2a) type of site with different values of the parameter u. Figure VI-3 shows the coordinates of the atoms in the orthorhombic cell drawn using the approximation  $\approx \frac{1}{2}$  for 0.490 and 0.510 and  $\approx \frac{1}{4}$  for 0.253 and 0.237.

A comparison of Eqs. VI-1 to VI-3 indicates that the transformation from cubic to tetragonal involves only shifts in the z coordinates of atoms, while the orthorhombic phrase differs from the cubic one only through shifts in atom positions within x,y planes (see Table VI-1).

#### 4. Atom Arrangements

The ionic radii of  $Ba^{2+}$  (1.34 Å) and  $O^{2-}$  (1.32 Å) are almost the same, and together they form a face-centered cubic (fcc) close-packed lattice with the smaller  $Ti^{4+}$  ions (0.68 Å) located in octahedral holes. The octahedral holes of a close-packed oxygen lattice have a radius of 0.545 Å, and if these holes were empty the lattice parameter would be a=3.73, as shown on Fig. VI-4a. If each

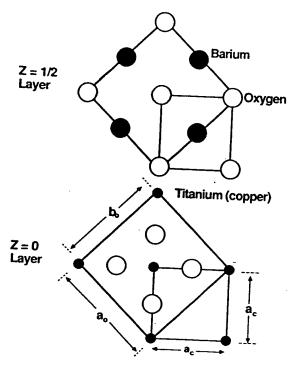


Fig. VI-3. Atom positions of perovskite when the monomolecular tetragonal unit cell is expanded to the bimolecular orthorhombic cell with new axes at 45° with respect to the old ones.

titanium were to move the surrounding oxygens apart to its ionic radius when occupying the hole, as shown on Fig. VI-4b. the lattice parameter a would be 4.00 Å. The observed cubic (a=4.012 Å) and tetragonal (a=3.995 Å, c=4.034 Å) lattice parameters are close to these values, indicating a pushing apart of the oxygens. The tetragonal distortion illustrated on Fig. VI-2 and the orthorhombic distortion of Eq. (VI-3) constitute attempts to achieve this through an enlarged but distorted octahedral site. This same mechanism is operative in the oxide superconductors.

### C. BARIUM-LEAD-BISMUTH OXIDE

In 1983 Mattheiss and Hamann referred to the 1975 "discovery by Sleight et al. of high temperature superconductivity" of the compound  $BaPb_{1-x}Bi_xO_3$  in the composition range  $0.05 \le x \le 0.3$  with  $T_c$  up to 13 K (Matt7, Sleig). Many consider this system, which disproportionates 2  $Bi^{4+} \rightarrow Bi^{3+} + Bi^{5+}$  in going from the metallic to the semiconducting state, as a predecessor to the LaSrCuO system.

Fig. dral the h para:

C: BaPt and 6.136  $\beta = 6$  and 1 4.35 at roc tetra;

TABLE VI-3. Atom Positions of Regular and Alternate La<sub>2</sub>CuO<sub>4</sub> Structure, Both of Which Correspond to Space Group 14/mmm, D<sub>4h</sub><sup>17a</sup>

						46						
<b>6</b> .			Regul	ar St	ructi	ıre		Alterna	ternate Structure			
Complex	Ideal z	Atom	Site	x	у	z	Atom	Site	x		z	
CuO <sub>2</sub>	•	(0(1)	4c	1/2	0	1	O(1)	4c	1/2	0	1	
CuO <sub>2</sub>	1	<b>O(1)</b>	4c	0	1/2	1	O(1)	4c	Ő	1/2	1	
		Cu	2a	0	0	1	Cu	2a	0	0	î	
OLa	$\frac{5}{6} = 0.833$	{ La (O(2)	4e 4e	1 2 0	1 2 0	0.862 0.818	La	4e	1 2	1 2	0.862	
							O(2) O(2)	4d 4d	0 1 2	1 2 0	, <u>1</u>	
LaO	$\frac{2}{3} = 0.667$	O(2) La	4e 4e	1 2 0	1 2 0	0.682				-	4	
		(O(1)	4c	Ö	1/2	0.638	La	4e	0	0	0.638	
O <sub>2</sub> Cu	1/2	O(1)	4c		0	1 1 1	O(1)	4c	0	1 2	1/2	
	۶.	(Cu	2a	1	Ţ	1 1 2 2	O(1)	4c	1 2 1 2	0	1/2	
LaO	$\frac{1}{3} = 0.333$	La O(2)	4e 4e	1 2 1 2 0 1 2	1/2 0 1/2	0.362 $0.318$	Cu La	2a 4e	<u>\$</u>	0	0.362	
				-	•		O(2)	4d	1 2	0	1	
OLa	$\frac{1}{6} = 0.167$	O(2)	4e	0	0	0.182	O(2)	4d	0	1 2	1 4 1 4	
<b>-</b>	= - U.16/	La	4e		1/2	0.138	La	4.	,			
	(	O(1)	4c	1 1 2	Õ	0.150	O(1)	4e 4c	1 1 2	1 2	0.138	
CuO₂	0 }	O(1)	4c	Ó.	1 2	ő	O(1)	4c 4c	2 0	0	0	
	(	Cu	2a	0	ó	ŏ	Cu	40 2a	0	$\frac{1}{2}$	0	

<sup>\*</sup>Superconducting compounds crystallize in the regular structure (Oguch; see also Onoda). The ideal z values in column 2 are for the prototype perovskite.

constants for tetragonal LaSrCuO superconductors with various values of x, y, and  $\delta$  in the formula  $(La_{1-x}Sr_x)_{2-y}CuO_{4-\delta}$ .

## 2. Alternate Tetragonal Form

In the previous section we discussed the tetragonal structure which is adopted by LaSrCuO superconductors. It has a variant (Hutir, Oguch) called the Nd<sub>2</sub>CuO<sub>4</sub> structure in which the oxygens O(2) are in special sites (4d) instead of the general (4e) sites in the same space group, corresponding to

O(2) (4d) 
$$0,\frac{1}{2},\frac{1}{4};\frac{1}{2},0,\frac{1}{4};\frac{1}{2},0,\frac{3}{4};0,\frac{1}{2},\frac{3}{4}$$
 (VI-7)

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The remaining atoms are in the positions given by Eq. (VI-6) and listed in Table VI-3, and the unit cell is sketched on the right-hand side of Fig. VI-5. This structure tends to be unstable relative to its  $K_2NiF_4$  counterpart, and is not known to superconduct.

tetragonal phase, and the metal-to-insulator transition occurs at the tetragonal-to-orthorhombic phase boundary  $x \approx 0.35$  (Matt7, Slei1).

## D. PEROVSKITE-TYPE SUPERCONDUCTING STRUCTURES

In their first report on high-temperature superconductors Bednorz and Müller referred to their samples as "metallic, oxygen deficient . . . perovskite like mixed valent copper compounds." Subsequent work has confirmed that the new superconductors do indeed have these characteristics. In this section we will comment on their perovskite-like aspects.

#### 1. Atom Sizes

In the oxide superconductors Cu replaces the Ti<sup>4+</sup> ions (0.68 Å) of perovskite, and in most cases retains the CuO<sub>2</sub> layering with two oxygens per copper in the layer. Other cationic replacements tend to be Bi, Ca, La, Sr, Tl, and Y for the larger Ba, forming "layers" containing only one oxygen or none per cation. We see from the following list of ionic radii

that there are four size groups, with all other cations significantly smaller then the Ba of perovskite. The common feature of  $CuO_2$  layers that are planar or close to planar establishes a fairly uniform lattice size in the a,b plane. The parameters of the compounds LaSrCuO (a=b=3.77 Å), YBaCuO (a=3.83 Å, b=3.89 Å), BiSrCaCuO (a=b=3.82 Å), and TIBaCaCuO (a=b=3.86 Å) are all between the ideal fcc oxygen lattice value of 3.73 Å and the perovskite one of 4.01 Å.

Table VI-2 gives the ionic radii of the positively charged ions of various elements of the periodic table. These radii are useful for estimating changes in lattice constant when ionic substitutions are made in existing structures. They also provide some insight into which types of substitutions will be most favorable.

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Bednorz and Müller
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8 Å) of perovskite, ns per copper in the r, Tl, and Y for the ione per cation. We

(VI-4)

cantly smaller then t are planar or close plane. The parame-(a = 3.83 Å, b =(a = b = 3.86 Å)and the perovskite

ions of various eleting changes in latructures. They also e most favorable.

TABLE VI-2. Ionic Radii in Angstroms of Selected Elements for Various Positive Charge States"

Z	Element	+1	+2	+3	+4	+5	+6
			Alka	ıli			
3	Li	0.68					
11	Na	0.97					
19	K	1.33	•				
37	Rь	1.47					
55	Cs	1.67					
			Alkaline	earths			
4	Be	0.44	0.35				
12	Mg	0.82	0.66			-	
20	Ca	1.18	0.99				
38	Sr		1.12				
56	Ba	1.53	1.34				
	•		Group	III			
5	В	0.35		0.23			
13	Al			0.51			
31	Ga	0.81		0.62			
49	ln			0.81			
81	TI	1.47		0.95			
			Group	IV			
6	C				0.16		
14	Si	0.65			0.42		
32	Ge		0.73		0.53		
50	Sn		0.93		0.71		
82	Pb		1.20		0.84		
			Group	, V			
15	P			0.44		0.35	
33	As			0.58		0.46	
51	Sb	0.89		0.76		0.62	
83	Bi	0.98		0.96		0.74	
			Chalcogo	enides			
16	S				0.37		0.30
34	Se	0.66	~		0.50		0.42
52	Te	0.82			0.70		0.56
		Fir:	st transition	series 13d°	,		
21	Sc			0.81			
22	Ti	0.96	0.94	0.76	0.68		
23	v		0.88	0.74	0.63	0.59	
24	Cr	0.81	0.89	0.63			0.52
25	Mn		0.80	0.66	0.60		

Z	Element	+1	+2	+3	+4	+5	+6
26	Fe		0.74	0.64			
27	Co		0.72	0.63			
28	Ni		0.69				
29	Cu	0.96	0.72				
30	Zn	0.88	0.74				
		Seco	nd transitio	n series (4d	·*)		
39	<b>Y</b>			0.94		•	
40	Zr	1.09			0.79		
41	Nb	1.00			0.74	0.69	
42	Мо	0.93			0.70		0.6
43	Tc						
44	Ru				0.67		
45	Rh			0.68			
46	Pd		0.80		0.65		
47	Ag	1.26	0.89				
48	Cď	1.14	0.97				
		Thi	rd transition	n series (5d'	y		
72	Hf				0.78		
73	Ta					0.68	
74	W				0.70		0.62
75	Re				0.72		
76	Os				0.88		0.69
77	lr				0.68		
78	Pt		0.80		0.65		
79	Au	1.37		0.85			
80	Hg	1.27	1.10				
			Rare eart	hs (4f <sup>n</sup> )			
57	La	1.39		1.14			
58	Се	1.27		1.07	0.94		
59	Pr			1.06	0.92		
60	Nd	•		1.04			
61	Pm			1.06			
62	Sm -	•		1.00			
63	Eu			0.98			
64	Gd			0.62			
65	Тъ			0.93	0.81		
66	Dу			0.92			
67	Ho			0.91			
68	Er			0.89			
69	Tm			0.87			
70	Yb			0.86			
71	Lu			0.85			

<sup>\*</sup>Three anion radii are 1.32 for O<sup>2-</sup>, 1.33 for F<sup>-</sup>, and 1.84 for S<sup>2-</sup> (Handbook of Chemistry and Physics).

0.69

0.68

0.62

0.62

0.69

Three and four fundamental fcc unit cells stack vertically to form the superconducting unit cells of YBaCuO and LaSrCuO, respectively, with some oxygens removed in the process. This causes the vertical height or c parameter of the unit cell to be less than that expected for the stacking of perovskite cells:

YBaCuO: 
$$c \approx 11.7 \text{ Å}, 3c_{fcc} = 11.19 \text{ Å}, 3c_{per} = 12.03 \text{ Å}$$
  
LaSrCuO:  $c \approx 13.18 \text{ Å}, 4c_{fcc} = 14.92 \text{ Å}, 4c_{per} = 16.04 \text{ Å}$  (VI-5)

Similar stackings occur in the BiSrCaCuO and TlBaCaCuO compounds.

#### E. LANTHANUM-COPPER OXIDE

2. Unit Cell Stacking

The structure of LaSrCuO,  $(La_{1-x}M_x)_2CuO_{4-\delta}$ , called the 21 structure, where M is usually Sr or Ba, is tetragonal in some cases and orthorhombic in others. We will describe the tetragonal case first and then the orthorhombic distortion of it. The structures will be described in terms of the prototype compound La<sub>2</sub>CuO<sub>4</sub> corresponding to  $x = \delta = 0$  in the above expression, keeping in mind that in the superconducting compounds themselves some of the La atoms are replaced by a divalent cation such as Sr or Ba. Since lanthanum has a charge of +3 and oxygen is -2, it follows that all of the copper is divalent (+2) when x = 0, and some becomes trivalent for x > 0.

The compound La<sub>2</sub>CuO<sub>4</sub> itself is considered to be nonsuperconducting, but some investigators claim that it or portions of it do exhibit superconductivity, perhaps of a filimentary type (Beill, Coop1, Dvora, Gran1, Pick1, Shahe, Skelt, Skel1, Skel2).

#### 1. Tetragonal Form

The tetragonal LaSrCuO superconductors crystallize in what is called the  $K_2NiF_4$  structure with space group I4/mmm,  $D_{4h}^{17}$  and two formula units per unit cell (e.g., Burns, Coll1, Hirot, Mossz, Onoda; Wyck3, p. 68). The copper atoms and one of the oxygen types O(1) are in special positions and the remaining atoms are all in general positions, with a single undetermined parameter associated with the z coordinate. The positions are

La (4e) 
$$0,0,u$$
;  $0,0,-u$ ;  $\frac{1}{2},\frac{1}{2},u+\frac{1}{2}$ ;  $\frac{1}{2},\frac{1}{2},-u+\frac{1}{2}$   
Cu (2a)  $0,0,0$ ;  $\frac{1}{2},\frac{1}{2},\frac{1}{2}$   
O(1) (4c)  $0,\frac{1}{2},0$ ;  $\frac{1}{2},0,0$ ;  $\frac{1}{2},0,\frac{1}{2}$ ;  $0,\frac{1}{2},\frac{1}{2}$   
O(2) (4e)  $0,0,v$ ;  $0,0,-v$ ;  $\frac{1}{2},\frac{1}{2},v+\frac{1}{2}$ ;  $\frac{1}{2},\frac{1}{2},-v+\frac{1}{2}$ 

with u=0.362 and v=0.182. Typical lattice dimensions are a=b=3.77 Å, c=13.18 Å. Table VI-3 gives more details on the atom positions and Fig. VI-5a provides a sketch of this 21 structure. Table VI-4 lists the measured lattice

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		]	Regula	r Strı	ıctur	Regular Structure					re
Complex	Ideal z	Atom	Site	x	у		Atom	Site	x	у	z
		(O(1)	4c	1 2	0	1	O(1)	4c	1 2	0	1
CuO <sub>2</sub>	1	O(1)	4c	Ó	12	1	O(1)	4c	0	1 2	1
CuOy	•	(Cu	2a	0	ō	1	Cu	2a	0	0	1
	_	(Ia	4e	Ť		0.862	La	4e	1 2	1/2	0.862
OLa	$\frac{5}{6} = 0.833$	{La O(2)	4e	1 2 0	1 2 0	0.818					
		(0(2)					O(2)	4d	0	$\frac{1}{2}$	3 4 3 4
							O(2)	4d	1 2	0	34
		(0(2)	4e	1 2	1 2	0.682					
LaO	$\frac{2}{3} = 0.667$	O(2) La	4e	Ô	ō	0.638	La	4e	0	0	0.638
		(0(1)	4c	0		1 2	O(1)	4c	0	1/2	1/2
O <sub>2</sub> Cu	$\frac{1}{2}$	<b>O(1)</b>	4c		1 2 0 1 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	1/2 1/2	O(1)	4c	1 1 1 2	1/2 0 1/2	1 1 2 1 2 1 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 2 1 2 1 2 1 2 1 2 1 2 1 2 1 2 1 2 1 2 1 2 1 2 1 2 1 2 1 2 1 2 1 2
Ozcu	2	(Cu	2a	1/2	1 2	1/2	Cu	2a	1/2	1/2	1/2
		La	4e	1 1 2 1 2	Ó	0.362	La	4e	0	0	0.362
LaO	$\frac{1}{3} = 0.333$	O(2)	4e	1 2	1/2	0.318					
		(-,-)		-			O(2)	4d	1/2	0	1 4 1 4
							O(2)	4d	0	0 1 2	14
	,	(O(2)	4e	0	0	0.182					
OLa	$\frac{1}{6} = 0.167$	La	4e		1 2	0.138	La	4e	1/2	1 2	0.138
		(O(1)	4c	1 1 2 0	Ó	0	O(1)	4c	1/2	0	0
CuO <sub>2</sub>	0	0(1)	4c	Ó	1 2	0	O(1)	4c	0	1 2	0
CuOy	Ū	(Cu	2a	0	Ó	0	Cu	2a	0	0	0

<sup>&</sup>quot;Superconducting compounds crystallize in the regular structure (Oguch; see also Onoda). The ideal z values in column 2 are for the prototype perovskite.

constants for tetragonal LaSrCuO superconductors with various values of x, y, and  $\delta$  in the formula  $(La_{1-x}Sr_x)_{2-y}CuO_{4-\delta}$ .

#### 2. Alternate Tetragonal Form

In the previous section we discussed the tetragonal structure which is adopted by LaSrCuO superconductors. It has a variant (Hutir, Oguch) called the Nd<sub>2</sub>CuO<sub>4</sub> structure in which the oxygens O(2) are in special sites (4d) instead of the general (4e) sites in the same space group, corresponding to

O(2) (4d) 
$$0, \frac{1}{2}, \frac{1}{4}; \frac{1}{2}, 0, \frac{1}{4}; \frac{1}{2}, 0, \frac{3}{4}; 0, \frac{1}{2}, \frac{3}{4}$$
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The remaining atoms are in the positions given by Eq. (VI-6) and listed in Table VI-3, and the unit cell is sketched on the right-hand side of Fig. VI-5. This structure tends to be unstable relative to its  $K_2NiF_4$  counterpart, and is not known to superconduct.

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1 2	0	1
0	0 1/2 0	1
0	0	1
1/2	1/2	0.862
0	ļ	34
0	0	2 4 2 4
0	0	0.638
0 0 1 1 2 1 2 0	1 2 0 1 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	1 1 2 1 2 1 2 1 2 1 2 2 1 2 2 2 2 2 2 2
1/2	0	1/2
1 2	1/2	1/2
0	0	0.362
1	.0	14
0	·0	1 1 4
1 2	1 2	0.138
1/2	Ō	0
ō	1 2 0 1 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	0
1 2 1 2 0 0 0	ō	0

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(VI-7)

and listed in Table . VI-5. This strucnd is not known to

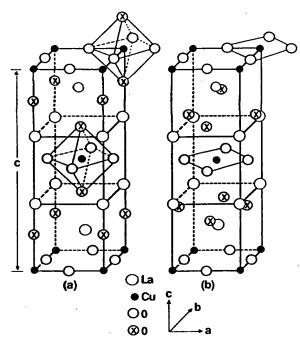


Fig. VI-5. Lanthanum copper oxide tetragonal unit cell. The regular cell (a) associated with the superconducting compounds is shown on the left and the alternative one (b) is on the right (Oguch; see also Ohba1). The oxygens denoted by  $\otimes$  have different positions in the two cells.

#### 3. Orthorhombic Form

The 21 orthorhombic LaSrCuO structure (Longo) is related to its tetragonal analogue given by Eq. (VI-6) in the same way that the orthorhombic perovskite structure (VI-3) is related to its tetragonal (VI-2) and cubic (VI-1) forms. This means that the orthorhombic basis directions are at 45° relative to the tetragonal ones, and the number of formula units in the cell is doubled. The situation is similar to that described by Fig. VI-3, with  $a = 5.363 \text{ Å} = 3.792\sqrt{2} \text{ Å}$ , b =5.409  $\dot{A} = 3.825\sqrt{2} \dot{A}$ ,  $c = 13.17 \dot{A}$ . Writing the a and b lattice parameters times  $\sqrt{2}$  compensates for the new choice of axes and shows that the orthorhombic values are close to the tetragonal a = 3.81 Å given earlier. There is also very little change in c. Table VI-5 lists the measured lattice constants for several orthorhombic compounds. The anisotropy factors ANIS

ANIS = 
$$\frac{100 |b - a|}{0.5 (b + a)}$$
 (VI-8)

listed in column 6 give the percentage deviation from tetragonality.

	meters <sup>b</sup>	Lattice Para	nai Structure	WICE 1011-80
Ref.	c (Å)	a = b(A)	x	Ŕ-M
Alige	12.68	3.828	0.4	
Skelt	13.168	3.782		Y-Ba
Yuzzz	13.2487	3.7817	0.05	La-Ba
Fujit	13.31	3.787	0.075	
Fujit	13.35	3.791	0.075	
Taral	13.211	3.7839	0.1	
Hidal	13.25	3.78 .	0.05	La-Sr
Taral	13.216	3.7784	0.05	
Decre	13.2	3.7793	0.063	
Tara	13.226	3.7771	0.075 0.075	
Shelt	13.234	3.776		
Brun	13.247	3.772	0.075	
Tara	13.232	3.7739	0.075	
Tara	13.23	3.7739	0.087	
Przys	13.2309	3.777	0.1	
Tara	13.242	3.7708	0.1	
Tara	13.247	3.7685	0.112	
Tara	13.255	3.7666	0.125	
Tara	13.259	3.7657	0.132 0.15	

\*The table is sorted by cations and then by increasing x, the dopant parameter (prepared by M. M. Rigney).

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The a and b lattice parameters were converted from measured values of  $a_0$ ,  $b_0$  of Fig. VI-3 through the expression  $a = a_0/\sqrt{2}$ ,  $b = b_0/\sqrt{2}$ .

Copper atoms and one of the oxygen types O(1) are in special positions; the remaining two atoms La and O(2) are in general positions with a single undetermined parameter associated with the z coordinate. The space group is Fmmm,  $D_{2h}^{23}$ , and the positions of the atoms are as follows:

La (8i) 
$$0,0,u$$
;  $0,\frac{1}{2},\frac{1}{2}+u$ ;  $\frac{1}{2},0,\frac{1}{2}+u$ ;  $\frac{1}{2},\frac{1}{2},u$ ;  $0,0,-u$ ;  $0,\frac{1}{2},\frac{1}{2}-u$ ;  $\frac{1}{2},0,\frac{1}{2}-u$ ;  $\frac{1}{2},\frac{1}{2},-u$   
Cu (4a)  $0,0,0$ ;  $0,\frac{1}{2},\frac{1}{2}$ ;  $\frac{1}{2},0,\frac{1}{2}$ ;  $\frac{1}{2},\frac{1}{2},0$   
O(1) (8e)  $\frac{1}{4},\frac{1}{4},0$ ;  $\frac{1}{4},\frac{3}{4},\frac{1}{2}$ ;  $\frac{3}{4},\frac{1}{4},\frac{1}{2}$ ;  $\frac{3}{4},\frac{3}{4},0$   
 $\frac{1}{4},\frac{1}{4},\frac{1}{2}$ ;  $\frac{1}{4},\frac{3}{4},0$ ;  $\frac{3}{4},\frac{1}{4},0$ ;  $\frac{3}{4},\frac{3}{4},\frac{1}{2}$   
O(2) (8i)  $0,0,v$ ; ... (same as La with  $v$  replacing  $u$ )

where the parameters u=0.362 and v=0.182 have the same values as in the tetragonal case presented above. Since u and v are the same and the lattice constants are so close to the tetragonal values, the sketch of the tetragonal unit cell in Fig. VI-5a applies here also. Another work (Hirot, see also Onoda) assigned

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Ref.
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Skelt
Yuzzz
Fujit
Fujit
Tara1
Hidak
Tara1
<b>Decro</b>
Tara1
Shelt
Brunz
Tarai
Taral
Przys
Taral
Taral
Taral
Tara1

epared by M. M.

Fig. VI-3 through

positions; the ingle undeter-

(VI-9)

u)

alues as in the he lattice congonal unit cell 10da) assigned

TABLE VI-5. Selected Lattice Parameters for (R<sub>1-x</sub>M<sub>x</sub>)<sub>2</sub>CuO<sub>4-5</sub> Type Superconductors with the Orthorhombic Structure<sup>2</sup>

		La	attice Paramet	ers	•	
R-M	x	a (Å)	b (Å)	c (Å)	ANIS	Ref. Fujit Shelt Onoda Hirot
La-Ba	0.02	3.786	3.811	13.17	0.66	Fujit
	0.075	3.786*	3.808*	13.257	0.58	Shelt
	0.075	3.798*	3.803*	13.234	0.13	Onoda
La-Ba	0.1	3.786*	3.824*	13.264	1.00	Hirot
La-Ca	0.075	3.772*	3.808*	13.168	0.95	Shelt

<sup>&</sup>quot;ANIS is the anisotropy factor 100|b-a|/0.5(b+a) (prepared by M. M. Rigney).

 $(\text{La}_{0.9}\text{Ba}_{0.1})_2\text{O}_4$  to the space group Pccm,  $D_{2h}^3$  with  $a=5.354=3.786\sqrt{2}$  Å,  $b=5.408=3.824\sqrt{2}$  Å, and c=13.264 Å.

#### 4. Phase Transition

The compounds  $(La_{1-x}M_x)_2CuO_4$  with M = Sr and Ba are orthorhombic at low temperatures and low M contents, and tetragonal otherwise, and superconductivity has been found on both sides of this transition (Baris, Bedn3, Birge, Dayzz, Dvora, Fujit, Gree1, Kangz, Koyam, Mihal, Paulz; see also Heldz). The prototype compound  $La_2CuO_4$  itself also exhibits the tetragonal-to-orthorhombic transition. The phase diagram of Fig. VI-6 shows the tetragonal, orthorhom-

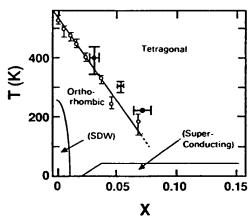


Fig. VI-6. Phase diagram showing data points along the tetragonal-to-orthorhombic transition line for  $(La_{1-x}Ba_x)_2CuO_{4-5}$  ( $\bigcirc$ , Fujit) and  $(La_{1-x}Sr_x)_2CuO_4$  ( $\bullet$ , Moret). The spin-density wave (SDW) and superconducting  $\bigcirc$  regions are indicated. These two compounds have about the same superconducting region.

<sup>&</sup>lt;sup>b</sup>The a and b lattice parameters were converted from the measured values of  $a_0$ ,  $b_0$  of Fig. VI-3 through the expressions  $a = a_0/\sqrt{2}$ ,  $b = b_0/\sqrt{2}$ .

86

bic, superconducting, and spin-density wave (SDW) regions for the barium compound (Fujit), and data points for the strontium compound (Moret, More8). An alternate phase diagram has been proposed (Ahar1). Alkaline metal contents much larger than those shown on the figure (e.g.,  $x \approx 0.5$ ) can be nonsuperconducting. The SDW region occurs below the minimum concentration for the onset of superconductivity. Another work (Geise) showed that LaSr(0.04) undergoes a structural phase transition between 180 and 300 K.

#### 5. Generation of LaSrCuO Structures

The LaSrCuO tetragonal structures may be visualized as being derived from four LaCuO<sub>3</sub> perovskite unit cells of the type illustrated in Fig. VI-1 stacked one above the other along the z or c axis. To generate La<sub>2</sub>CuO<sub>4</sub> in the K<sub>2</sub>NiF<sub>4</sub> structure the layers of CuO<sub>2</sub> atoms on the  $z=\frac{1}{4}$  and  $z=\frac{3}{4}$  levels of this four-cell stacking are removed, La and O are interchanged on two other layers, and the middle layer Cu atom is shifted from the edge to the center point  $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$  of the unit cell. Then the cell is compressed vertically from 14.9 to 13.2 Å (Table VI-4) to take up the space formerly occupied by the removed CuO<sub>2</sub> layers. Finally, the lanthanums along the c axis and the oxygens along the side edges are shifted vertically to accommodate the new atom arrangement.

To generate La<sub>2</sub>CuO<sub>4</sub> with the Nd<sub>2</sub>CuO<sub>4</sub> arrangement from this same four-cell stacking all of the oxygens on the vertical edges are removed, and two lanthanums are moved to edge sites. Copper is handled the same way as before, so

in both cases the generated structure lacks two CuO2 layers.

#### 6. Layering Scheme of LaSrCuO

When we described the LaSrCuO structures we left out what is perhaps their most important characteristic, namely, their layered aspect. Lanthanum copper oxide may be looked upon as consisting of Cu-O layers of square-planar coordinated copper ions with lanthanum and O(2)-type oxygen ions populating the spaces between the layers. These Cu-O layers are stacked equally spaced, perpendicular to the c axis, as shown in Fig. VI-7, and their oxygens are aligned along the c axis, as indicated by the vertical dotted line on the left side of the figure. The copper ions, on the other hand, are not aligned vertically, but rather alternate between (000) and  $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$  sites in adjacent layers, as illustrated in Figs. VI-5 and VI-7.

The copper is actually octahedrally coordinated with oxygen, but the Cu-O distance of 1.9 Å in the CuO<sub>2</sub> planes is much less than the vertical distance of 2.4 Å between copper and the oxygens above and below, as shown in Fig. VI-8. When the structure is distorted orthorhombically the Cu-O spacings in both the planes and the c direction remain quite close to their tetragonal counterparts.

The copper ions and the O(1)-type oxygens in the planes are both in special sites in the tetragonal and orthorhombic forms, in accordance with Eqs. (VI-6) and (VI-9), and as a result the plane is perfectly flat in both cases. When the

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Attachment D

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CLAIMS (CARBON LIKE)

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For further explanation, press the H key (for HELP) and then the ENTER key.

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CLAIMS (SILICON LIKE)

Your search request has found 5 PATENTS through Level 1. To DISPLAY these PATENTS press either the KWIC, FULL, CITE or SEGMTS key. To MODIFY your search request, press the M key (for MODFY) and then the ENTER key.

For further explanation, press the H key (for HELP) and then the ENTER key.

AHachment H

CLAIMS (NITROGEN-LIKE)

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To MODIFY your search request, press the M key (for MODFY) and then the ENTER key.

For further explanation, press the H key (for HELP) and then the ENTER key.

A Hackment I

CLAIMS (COPPER LIKE)

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For further explanation, press the H key (for HELP) and then the ENTER key.

A Hach ment J

### Attachment M

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FOCUS - 1 OF 23 PATENTS

5,501,808

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'n

**Mar. 26, 1996** 

Crystalline-like transition metal material

INVENTOR: Patalano, Philip, 521 Lauiki St. #6, Honolulu, Hawaii 96826

transition metal acetylides, can obtain high molecular weights. However, the ... polymers. The compounds of the present invention, like the linear compounds of this invention are two-dimensional and three-dimensional in structure with many compounds being ceramic-like. Compounds of the present invention can be produced by reacting anhydrous transition metal halide with alkali metal and/or alkaline earth metal acetylides (C2<2 - > = acetylide)in an inert atmosphere following ...

FOCUS - 2 OF 23 PATENTS

5,490,977

Feb. 13, 1996

Removal of CO, hydrocarbons and NO x with catalyst containing platinum rhodium

INVENTOR: Wan, Chung-Zong, Somerset, New Jersey Tauster, Samuel J., Englishtown, New Jersey Rabinowitz, Harold N., Upper Montclair, New Jersey

catalytic material is dispersed as a coating on the carrier, specifically, on the walls of the gas flow passages thereof. Such carriers are normally made of a refractory, ceramic-like material such as cordierite, mullite, alumina, or any other suitable refractory material; they may also be made of a refractory metal such as stainless steel or other suitable corrosion-resistant, iron based

The ...

FOCUS - 3 OF 23 PATENTS

5,434,125

<=2> ...GET 1st DRAWING SHEET OF 1

Jul. 18, 1995

PAGE

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Rare earth oxide superconducting material and process for producing the same

INVENTOR: Ogawa, Naoyuki, Anjo, Japan Sakai, Takenobu, Susono, Japan Hirabayashi, Izumi, Nagoya, Japan

DETDESC:

above-mentioned mixed powder on a substrate made of a metal, a ceramic or the ... material of rare earth oxide superconducting material. It is also like to form a layer of rare earth oxide superconducting material on the possible to obtain a shaped material by spray- or powder-coating the substrate.

desired rare earth oxide superconducting material, used in the present invention The temperature equal to or higher than the incongruent melting point of

FOCUS - 4 OF 23 PATENTS

5,395,821

<=2> GET 1st DRAWING SHEET OF 3

Mar. 7, 1995

Method of producing Pb-stabilized superconductor precursors and method of producing superconductor articles therefrom

INVENTOR: Kroeger, Donald M., Knoxville, Tennessee Hsu, Huey S., Knoxville, Tennessee Brynestad, Jorulf, Oak Ridge, Tennessee

MILE

... transition temperature above 770 K. thereby allowing liquid nitrogen to be used as the cryogenic material. The high temperature superconducting materials generally consist of metal production, the metal oxides are mixed together as solids and heated at sintering temperatures of 7000 C. to 11000 C. The sintered material is then oxides bonded together to form a ceramic-like structure. In one method of reground and reheated. The material is pressed into pellets and ....

FOCUS - 5 OF 23 PATENTS

5,378,345

<=2> GET 1st DRAWING SHEET OF 6

Jan. 3, 1995

Ceramic solid electrolyte-based electrochemical oxygen

### concentrator cell

INVENTOR: Taylor, Dale M., Salt Lake City, Utah Joshi, Ashok V., Salt Lake City, Utah

DETDESC:

... ingredients may be incorporated to enhance the structural properties of the electrolyte. For example, minor quantities of ZrO2, HfO2 and the like may be utilized as well as minor amounts of alumina, mullite and like ceramic oxides to enhance sintering or structural properties.

The ionic conductivity (mobility of the oxygen ion) of the ceria electrolytes of this invention is significantly better than zirconia or hafnia electrolytes, for example, or even certain ceria or bismuth oxide ... PAGE

FOCUS - 6 OF 23 PATENTS

5,376,625

<=2> GET 1st DRAWING SHEET OF 1

Dec. 27, 1994

Method of making thin film superconductor assembly

INVENTOR: McCune, Robert C., Birmingham, Michigan

DETDESC:

carbide, strontium titanate, aluminum oxide and aluminum nitride. Other suitable and ceramic-like materials well known to the skilled of the art. Exemplary such coating 40. Preferred materials include various commercially available ceramic materials include beryllium oxide, diamond or diamond-like thin films, silicon sood structural integrity with substrate 15 and with superconductor 30 and

FOCUS - 7 OF 23 PATENTS

5,348,797

<=2> GET 1st DRAWING SHEET OF 1

Sep. 20, 1994

Copper oxide coated substrates

INVENTOR: Clough, Thomas J., Santa Monica, California Grosvenor, Victor L., Topanga, California Pinsky, Naum, Thousand Oaks, California

SUM

... about 150 microns, extrudates, flakes, single fibers, fiber rovings,

e.g., catalyst supports, multi-channel monoliths, tubes, conduits and the like. Ceramic and metal fibers, especially continuous fibers, are particularly chopped fibers, fiber mats, porous substrates, irregularly shaped particles, useful substrates when the copper oxide coated substrate is to be used as The conditions at which each of the steps of the present process occur are effective to obtain the ...

FOCUS - 8 OF 23 PATENTS

5,338,722

Aug. 16, 1994

Method of forming superconducting oxide ceramic materials having high critical densities of superconducting current

INVENTOR: Takemura, Yasuhiko, Kanagawa, Japan

... ceramic material in the crucible is maintained for 12 hours, and naturally cooled in order to complete the formation procedure.

comprised oxide superconducting plate-like ceramic crystals of 10 microns diameter. The critical density of superconducting current was measured to In accordance with experiments, the superconducting ceramic material 11000 A/cm<2> The increase of the oxygen partial pressure can be carried out at once by ... PAGE 9

FOCUS - 9 OF 23 PATENTS

5,292,716

Mar. 8, 1994

Oxide superconducting material and process for producing the

INVENTOR: Sakai, Hitoshi, Komaki, Japan Yoshida, Hitoshi, Okazaki, Japan Baba, Hideyuki, Nagoya, Japan Yoshida, Manabu, Aichi, Japan

DETDESC:

above-mentioned mixed powder on a substrate made of a metal, a ceramic or the ... material of rare earth oxide superconducting material. It is also possible to obtain a shaped material by spray- or powder-coating the like to form a layer of rare earth oxide superconducting material on the substrate

PAGE

10

5,279,852

GET 1st DRAWING SHEET OF 1

Jan. 18, 1994

Process for coating a substrate with copper oxide and uses for coated substrates

INVENTOR: Clough, Thomas J., Santa Monica, California Grosvenor, Victor L., Topanga, California Pinsky, Naum, Thousand Oaks, California

SOM:

like. Ceramic and metal fibers, especially continuous fibers, are particularly chopped fibers, fiber mats, porous substrates, irregularly shaped particles, e.g., catalyst supports, multi-channel monoliths, tubes, conduits and the ... about 150 microns, extrudates, flakes, single fibers, fiber rovings useful substrates when the copper oxide coated substrate is to be used as superconductor.

The conditions at which each of the steps of the present process occur are effective to obtain the ...

FOCUS - 11 OF 23 PATENTS

5,272,132

GET 1st DRAWING SHEET OF

Dec. 21, 1993

Apparatus comprising a ceramic superconductive body and method for producing such a body

INVENTOR: Gyorgy, Ernst M., Madison, New Jersey Johnson, Jr., David W., Pluckemin, New Jersey

SUM:

... used by these workers as being of the "Wayne State University type"

variety of apparatus including power transmission lines, rotating machinery such Among the techniques for producing the green bodies are extrusion, screen printing, tape casting, and slip casting. The inventive filamentary and sheet-like ceramic superconductive bodies can be advantageously used in a as electrical generators, magnets such as may be used in ...

H

DETDESC

limited. We believe that the techniques disclosed herein can, either directly or ... referred to co-assigned U.S. patent application, the invention is not so relatively small dimension, generally in the approximate range 5 or 10 mu m with obvious changes, be used in general in the manufacture of filamentary and/or sheet-like ceramic oxidic superconductive bodies having at least one

Such bodies not only are of substantial technological significance but their

ambient environment with greater partial pressure of 02 than that of air (0.2 ... properties. For most favorable results, the material is fired in an

A significant aspect of the invention is the formation of a filamentary or sheet-like ceramic superconductive body. In general, known techniques can be used to form the given body. These include extrusion, screen printing, tape casting, and slip casting.

The starting materials for each of these processes ...

FOCUS - 12 OF 23 PATENTS

5,254,519

Oct. 19, 1993

Catalyst composition containing platinum and rhodium components

Rabinowitz, Harold N., Upper Montclair, New Jersey INVENTOR: Wan, Chung-Zong, Somerset, New Jersey Tauster, Samuel J., Englishtown, New Jersey

DETDESC:

catalytic material is dispersed as a coating on the carrier, specifically, on the walls of the gas flow passages thereof. Such carriers are normally made of a other suitable refractory material; they may also be made of a refractory metal ... continuous and open-ended gas flow passages extending therethrough. The refractory, ceramic-like material such as cordierite, mullite, alumina, or any such as stainless steel or other suitable corrosion-resistant, iron based alloys.

FOCUS - 13 OF 23 PATENTS

5,183,799

GET 1st DRAWING SHEET OF 16

13 PAGE

Feb. 2, 1993

Superconducting materials including La-Sr-Nb-O, Y-Ba-Nb-O, La-Sr-Nb-Cu-O, and Y-Ba-Nb-Cu-O

INVENTOR: Ogushi, Tetsuya, Kagoshima, Japan Hakuraku, Yoshinori, Kagoshima, Japan

Ogata, Hisanao, Ibraki, Japan

... laminating this superconducting material with other films of electrical insulating material. It is preferable to laminate a plurality of film-like layers alternately, respectively. Further, it is preferable to use as an insulating material a perovskite-like ceramic of the same series. Further, in the above-mentioned formulae (1) and (2), a total of valence number (p) of L, A and M, or L, A, M and Cu, and the valence number y of oxygen

FOCUS - 14 OF 23 PATENTS

5,145,833

Sep. 8, 1992

Method for producing ceramic bodies

INVENTOR: Prunier, Jr., Arthur R., Midland, Michigan Spangenberg, Stanley F., Midland, Michigan Wijeyesekera, Sunil, Midland, Michigan

SUM:

... as to retain its configuration and removed from the casting mold.

The pressure-transmitting medium includes a rigid interconnected skeletal structure which is collapsible when a predetermined force is applied. The skeletal structure may be of a ceramic-like material which is rigid and retains its configuration, but which may be broken up, crushed, fractionated or caused to flow at a predetermined relatively minimal force. The skeletal structure is defined by the ceramic material being ...
FOCUS - 15 OF 23 PATENTS

5,132,283

GET 1st DRAWING SHEET OF

Jul. 21, 1992

Thin film superconductor assembly and method of making the

INVENTOR: McCune, Robert C., Birmingham, Michigan

carbide, strontium titanate, aluminum oxide and aluminum nitride. Other suitable and ceramic-like materials well known to the skilled of the art. Exemplary such coating 40. Preferred materials include various commercially available ceramic materials include beryllium oxide, diamond or diamond-like thin films, silicon ... 20 can be formed of any suitable dielectric material which can achieve good structural integrity with substrate 15 and with superconductor 30 and

16

FOCUS - 16 OF 23 PATENTS

5,057,483

Oct. 15, 1991

Catalyst composition containing segregated platinum and rhodium components

INVENTOR: Wan, Chung-Zong, Somerset, New Jersey

DETDESC:

specifically, on the walls of the gas flow passages thereof. Such carriers are therethrough. The catalytic material is dispersed as a coating on the carrier, normally made of a refractory, ceramic-like material such as cordierite, mullite, alumina, or any other suitable refractory material; they may also be ... parallel, continuous and openended gas flow passages extending made of a refractory metal such as stainless steel or other suitable corrosion-resistant, iron based alloys.

FOCUS - 17 OF 23 PATENTS

5,049,452

Sep. 17, 1991

Target member used for formation of superconducting film

INVENTOR: Takeshita, Takuo, Saitama, Japan

Sugihara, Tadashi, Saitama, Japan

SUM:

... cm<2 > for 1 to 4 hours. The target is fabricated in this manner.

R-A-Cu-O system containing the copper oxide (which is hereinbelow represented by CuO) equal to or less than 20% by volume. The target thus fabricated is mainly formed of a superconducting ceramic material in the R-A-Cu-0 system or of a substance like the ceramic in the

PAGE

However, the composition of the target may be transferred to the composition of the thin film ...

18

PAGE

FOCUS - 18 OF 23 PATENTS

4,975,413

Dec. 4, 1990

Superconductor-coated carbon fiber composites

INVENTOR: Satek, Larry C., Wheaton, Illinois Bennett, William F., Hartsdale, New York Schulz, David A., Fairview Park, Ohio

uses enormously improved. However, because these new mixed-oxide superconductors are brittle, ceramic-like materials, they do not lend themselves easily to fabrication in the form of high strength, wire-type geometries, a requirement for many important uses to which superconductors have been put in the past. ... lower superconducting-transition-temperature superconductors is large enough that many new uses for superconductors now can be devised and present These uses largely ...

FOCUS - 19 OF 23 PATENTS

4,949,702

GET 1st DRAWING SHEET OF 4

Aug. 21, 1990

Self-heating container

INVENTOR: Suzuki, Ryoichi, Yokohama, Japan Kawabata, Choji, Tatebayashi, Japan Takeuchi, Akira, Fukaya, Japan Kuwahara, Motoo, Kamaishi, Japan Yamauchi, Kunio, Hikone, Japan Maiya, Mitsuo, Tokyo, Japan Ando, Koki, Tokyo, Japan

DETDESC

... DRAWINGS

In FIG. 1, the container includes a cylindrical can or metal casing 20 with its outer side surface surrounded by a heat-insulating cover 22 of paper, plastic, cloth, ceramic or the like. The cover 22 reduces the heat radiation from the can 20, and facilitates the handling of the container.

The can 20 contains a heater 24 near the bottom thereof, the heater including a cylindrical inverted cup-shaped ...

FOCUS - 20 OF 23 PATENTS

GET 1st DRAWING SHEET OF <=2>

May 22, 1990

Method of methanol production

INVENTOR: McShea, III, William T., Martinsville, New Jersey Yarrington, Robert M., Westfield, New Jersey

DETDESC:

resistance, and though not always, low thermal conductivity. Two general types of material of construction for such carriers are known. One is a ceramic-like porous material comprised of one or more metal oxides, for example; alumina, alumina-silica, alumina-silica-titania, mullite, cordierite, zirconia, ... exhibits a low thermal coefficient of expansion, thermal shock zirconia-spinel, zirconia-mullite, silicon carbide, etc. ... ... extending therethrough. The sheets and corrugations are sized to provide the desired number of gas flow passages, which may range, typically, from about 200 to 1,200 per square inch of end face of the tubular roll.

have a relatively low surface area with respect to catalyst support requirements alumina-silica-titania are somewhat porous and rough-textured, they nonetheless Although the ceramic-like metal oxide materials such as cordierite or and, of course, a stainless

FOCUS - 21 OF 23 PATENTS

4,320,418

σ GET 1st DRAWING SHEET OF

Mar. 16, 1982

Large area display

INVENTOR: Pavliscak, Thomas J., 2 S 454 Seneca Dr., Wheaton, Illinois 60187

DETDESC:

materials to the surface. Therefore, the electrodes are preferably deposited on ... substrate surface inhibits the permanent adherence of some electrode that glass substrate surface which is free from tin or tin oxide.

ceramic-like material containing one or more oxides such as aluminum oxide, silicon oxide, titanium oxide, zirconium oxide, magnesium oxide, lead oxide, and In another mode of this invention, the substrate is of a ceramic or so forth

Since visible light generated by the monolithic ...

FOCUS - 22 OF 23 PATENTS

4,233,623

<=2> GET 1st DRAWING SHEET OF 7

Nov. 11, 1980

Television display

INVENTOR: Pavliscak, Thomas J., 2 S. 454 Seneca Dr., Wheaton, Illinois 60187

DETDESC:

0 materials to the surface. Therefore, the electrodes are preferably deposited ... substrate surface inhibits the permanent adherence of some electrode that glass substrate surface which is free from tin or tin oxide.

silicon oxide, titanium oxide, zirconium oxide, magnesium oxide, lead oxide, and In another mode of this invention, the substrate is of a ceramic or ceramic-like material containing one or more oxides such as aluminum oxide,

Since visible light generated by the monolithic ...

FOCUS - 23 OF 23 PATENTS

3,996,447

GET 1st DRAWING SHEET OF 2

Dec. 7, 1976

PTC resistance heater

INVENTOR: Bouffard, Michael L., Pawtucket, Rhode Island Grant, John L., Mansfield, Massachusetts

An electrical heater device includes a disc-like ceramic resistor element of a material of positive temperature coefficient of resistivity having contact surfaces formed on a broad opposite sides of the element. A pair of device terminals engage respective contact surfaces of the resistor ... \* 23 PAGES 386 LINES JOB 79720

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35)

### ATTACHMENT A

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From: lexis-nexis@prod.lexis-nexis.com (LEXIS(R)/NEXIS(R) Print Delivery)
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LEVEL 1 - 1 OF 225 PATENTS

5,828,093

# <=2> GET 1st DRAWING SHEET OF 3

Oct. 27, 1998

Ceramic capacitor and semiconductor device in which the ceramic capacitor is mounted

INVENTOR: Naito, Yasuyuki, Kyoto, Japan Sakabe, Yukio, Kyoto, Japan

- abe, lukio, kjoto, capain ... [\*1] by a gap and surrounding said first capacitor electrode.
- [\*2] 2. A ceramic capacitor according to claim 1, wherein said ceramic dielectric substrate comprises a substrate for an SrTi03 boundary layer type semiconductive capacitor.
- [\*3] 3. A ceramic capacitor according to claim 1, further comprising outer electrodes which are mainly made of at least one material selected from the group consisting of Au, Pt and Pd and formed on at ...
- ... [\*12] layer is formed on said second principal face of said substrate.
- [\*13] 13. A ceramic capacitor according to claim 5, wherein said ceramic dielectrife substrate comprises a substrate for an SrTiO3 boundary layer type semiconductive capacitor.
- [\*14] 14. A ceramic capacitor according to claim 5, wherein said first capacitor electrode formed on said first principal face of said ceramic substrate is divided into two parts.
- [\*15] 15. ...
- layers disposed respectively on said first and second capacitor ... [\*19] electrodes.
- $[\star 20]$  20. A ceramic capacitor according to claim 19, wherein said ceramic dielectric substrate comprises a substrate for an SrTi03 boundary layer type semiconductive capacitor.
- [\*21] 21. A ceramic capacitor according to claim 20, further comprising outer electrodes which are mainly made of at least one material selected from the group consisting of Au, Pt and Pd and formed on at ...
- ... [\*25] layer formed on said first principal face of said substrate.
- [\*26] 26. A ceramic capacitor according to claim 23, wherein said ceramic diejectric substrate comprises a substrate for an SrTi03 boundary layer type semiconductive capacitor.
- $[*27] \ 27.$  A ceramic capacitor according to claim 23, wherein said first capacitor electrode formed on said first principal face of said ceramic

2

Pat. No. 5828093, \*27

8] 28. ...

LEVEL 1 - 2 OF 225 PATENTS

5.793.76

=2> GET 1st DRAWING SHEET OF 21

1

Aug. 11, 1998

ATM communication device and ATM communication network system with terminal devices having uniquely assigned virtual channel identifiers

INVENTOR: Soda, Keiichi, Kanagawa, Japan Ichihashi, Tatsuki, Kanagawa, Japan Ushisako, Yukio, Kanagawa, Japan Kashima, Kazuyuki, Kanagawa, Japan Yokotani, Tetsuya, Kanagawa, Japan Hiramatsu, Koichi, Kanagawa, Japan Shibahara, Makoto, Hyogo, Japan

- ... [\*6] ATM cell transmission section of the first ATM terminal communication device to the other ATM terminal communication devices is stored into a segmentation and reassemble sublayer protocol data unit for an ATM adaptation layer type 3 or 4 in addition to the field of the virtual path identifier and the virtual channel identifier in the first ATM cell.
- [\*7] 7. The ATM communication network system of claim 6, wherein the segmentation and reassemble sublayer ...
- device to the other ATM terminal communication devices is stored into a common part convergence sublayer protocol data unit for an ATM adaptation layer type 3 or 4 in addition to the field of the virtual path identifier and the virtual channel identifier in the first ATM cell.
- 9. The ATM communication network system of claim 8, wherein the common part convergence ... [6\*]
- into a common part convergence sublayer protocol data unit for an ATM adaptation layer type 5 in addition to the field of the virtual path identifier and the ... [\*10] space to be changed and transmitted from the ATM cell transmission section to the other ATM terminal communication devices is stored virtual channel identifier in the first ATM cell.
- 11. The ATM communication network system of claim 10, wherein the common part convergence sublayer ... LEVEL 1 - 3 OF 225 PATENTS [\*11]

PAGE

5,774,665

### =2> GET 1st DRAWING SHEET OF (

Jun. 30, 1998

Asynchronous transfer mode LAN switching hub device using IEEE P1355 standard and its control method

INVENTOR: Jeong, Seong-Ho, Yusong-ku, Republic of Korea Kim, Jang-Kyung, Yusong-ku, Republic of Korea Chong, Il-Young, Seo-ku, Republic of Korea

function, a bridging/relay layer for executing a bridging and relay function, an PAGE physical layer for directly connecting with the ATM network, an ATM layer for executing ATM layer for a AAL (ATM Adaptation Layer) type layer for an ATM adaptation layer) type layer for executing an ATM adaptation layer for executing an ATM adaptable function, a LAN emulation layer for executing LAN emulation

LEVEL 1 - 4 OF 225 PATENTS

5,764,658

<=2> GET 1st DRAWING SHEET OF 36

Jun. 9, 1998

Data receiving apparatus and method

INVENTOR: Sekiguchi, Shun-ichi, Kanagawa, Japan Murakami, Tokumichi, Kanagawa, Japan Kato, Yoshiaki, Kanagawa, Japan sequence, which are organized in a hierarchy including a digital coded bit each layer, which are organized in a hierarchy including a plurality of layers, each layer, having associated therewith one of a plurality of layer types, wherein at least one layer of a high-order layer type is composed of at least one layer of a lower-order layer type, each of the data blocks corresponding to one of the layers and including a start code which identifies the layer type of the data block, the apparatus comprising: a layer memory for storing data indicating the layer type corresponding to the last start code received in the bit stream, the layer memory having a last layer type output, a first expected start code selector coupled to receive the last layer type output from the layer memory, the first expected start code selector selecting and generating a first expected start code selector output identifying a set of expected start codes based on the last layer type received, wherein start codes of data blocks in the high-order layers are identified as expected start codes before start codes of data blocks in the lower-order layers;

start code

... [\*1] detected start code, if one of the expected start codes is similar to data in the bit stream, the start code detector having a start code detector output which identifies the detected start code and its corresponding layer

means responsive to the start code detector output for updating the layer memory with data indicating the layer type corresponding to the detected start

a block data decoder, responsive to the detected start code output from the start code detector, for decoding the data block corresponding to the detected start code.

[\*2] 2. An

sequence, which are organized in a hierarchy including a plurality of layers, each layer having associated therewith one of a plurality of layer types, wherein at least one layer of a high-order layer type is composed of at least one layer of a lower-order layer type, each of the data blocks corresponding to one of the layers and including a start code which identifies the layer of the data block, the method comprising the steps of:

Pat. No. 5764658, \*12 bit stream of data blocks, each comprising a digital coded bit ... [\*12]

storing data indicating the layer corresponding to the last start code ...

... [\*12] a set of expected start codes based upon the data indicating the layer wherein said set of expected start codes lists start codes of data blocks in layers of said high-order layer type before start codes of data blocks in layers of said lower-order layer type;

continuously comparing data in the bit stream with the set of expected start

selecting one of the expected start codes as a detected start code based on the comparison, if one of the expected start ...
LEVEL 1 - 5 OF 225 PATENTS

5,720,851

GET 1st DRAWING SHEET OF

Feb. 24, 1998

Method and arrangement for producing a foam-formed fibre or paper web

INVENTOR: Reiner, Lennart, Matfors, Sweden

plurality of dispersion vessels. ... [\*11]

PAGE

[\*12] 12. Arrangement according to claim 11, wherein the different fibre types are metered separately from the dispersion vessels up to an inlet box of the paper machine, said inlet box being of multi-layer type.

[\*13] [#13] [#13] Method according to claim 1, wherein the foam-formed fibre web inciudes a paper web [\*14] 14. Method according to claim 1, wherein the foamed fibre dispersion is formed by dispersing natural ...

LEVEL 1 - 6 OF 225 PATENTS

5,715,250

GET 1st DRAWING SHEET OF 4

Feb. 3, 1998

ATM-lan connection apparatus of a small scale capable of connecting terminals of different protocol standards and ATM-lan including the ATM-lan connection apparatus

INVENTOR: Watanabe, Ayumi, Tokyo, Japan

... [\*1] first ATM terminal for receiving as a first reception cell a first ATM cell supplied from said first ATM terminal;

a first AAL5-SAR (ATM Adaptation Layer type 5-Segmentation And Reassembly) section for reassembling said first reception cell into a first AAL5 packet to output said first AAL5 packet as a first LAN emulation frame;

:

... [\*3] first ATM terminal for receiving as a first reception cell a first ATM cell supplied from said first ATM terminal;

a first AAL5-SAR (ATM Adaptation Layer type 5-Segmentation And Reassembly) section for reassembling said first reception cell into a first AAL5 packet to output said first AAL5 packet as a first LAN emulation frame;

6

LEVEL 1 - 7 OF 225 PATENTS

5,714,403

GET 1st DRAWING SHEET OF

Feb. 3, 1998

1

Process for producing a matrix of "all optical" vertically-structured quantum well components

∞ PAGE

INVENTOR: Nissim, Yves, Paris, France Bensoussan, Marcel, Boulogne, France Oudar, Jean-Louis, Chatenay Malabry, France Rao, Elchuri, Issy Les Moulineaux, France

- ... [ $\star$ 2] in that the quantum well layer is a binary, ternary or quaternary GaAs or InP-based III-V semiconductor.
- [\*3] 3. A process according to claim 2, characterized in that the quantum well active layer is of the type GaAs/Ga[1-x]Al[x]As with 0 < -x < 1, or GaAs/Ga[1-x]In[x]As with 0 < x < 1 or InP/In[x]Ga[1-...
- ... [\*3] In[x]Ga[1-x]As[y]P[1-y ]with 0 </= x </= 1 and with 0 </= y </= 1.
- [\*4] 4. A process according to claim 1, characterized in that the positive layer is of type Si[x]N[y] or possibly Si0[x]N[y], with y, in the latter case, being small enough to enable the Si0[x]N[y] based layer to behave as a positive layer.
- [\*5] 5. A process according to claim 1, characterized in that the negative layer is of type Si[x]N[y] or possibly Si0[x]N[y], with y being small enough to enable the Si0[x]N[y] based layer to behave as a negative layer.

LEVEL 1 - 8 OF 225 PATENTS [**9**\*]

5,702,792

GET 1St DRAWING SHEET OF

Dec. 30, 1997

Optical recording medium

INVENTOR: Iida, Tetsuya, Tsurugashima, Japan Jinno, Satoshi, Tsurugashima, Japan Higuchi, Takanobu, Tsurugashima, Japan

What is claimed is:

- [\*1] 1. An optical recording medium of a multi-layer type comprising:
- a substrate
- a single or plural spacer layers each carrying pits and/or grooves;
- a single or plural reflective layers layered on the spacer layers; and

wherein said reflective layer is made of a ...

PAGE

... [\*1] OH groups and a surface of said reflecting layer furthest from said substrate contacting with said spacer layer is provided with a silane coupling treatment.

[\*2] 2. An optical recording medium of a multi-layer type comprising:

a substrate

a single or plural spacer layers each carrying pits and/or grooves;

a single or plural reflective layers layered on the spacer layers; and

wherein said reflective layer is made of one or ... LEVEL 1 - 9 OF 225 PATENTS

5,693,085

<=2> GET 1st DRAWING SHEET OF

Dec. 2, 1997

Stent with collagen

INVENTOR: Buirge, Andrew W., Minnepaolis, Minnesota Buscemi, Paul J., Long Lake, Minnesota Burmeister, Paul H., Maple Grove, Minnesota

... [\*21] combination of claim 20 wherein the collagen material includes Type I and Type IV layers.

[\*22] 22. The combination of claim 21 wherein the Type IV is SIS.

[\*23] 23. The combination of claim 22 wherein the Type IV is the innermost layer and the Type I layer includes a drug.

[\*24] 24. The combination of claim 17 wherein the stent is of variable diameter.

[\*25] 25. The combination of claim 24 wherein the stent is of the self-expanding type.

 $\left[ ^{\star}26 \right]$  26. The combination of claim 17 wherein the liner has ...

5,668,353

LEVEL 1 - 10 OF 225 PATENTS

<=2> GET 1st DRAWING SHEET OF

Sep. 16, 1997

PAGE

Input panel avoiding interference pattern and method of forming the same

٠ .

INVENTOR Matsuda, Genichi, Kawasaki, Japan Tanaka, Toshiaki, Kawasaki, Japan

What is claimed is:

[\*1] 1. An input panel of a resistance layer type comprising:

a first board having a first transparent conductive layer on one surface;

a second board having a second transparent conductive layer on one surface, said first board and said ...

- ... [\*3] as claimed in claim 1, wherein said second spacers are arranged at intervals from 0.5 mm to 20 mm, and said first spacers are arranged at smaller intervals
- [\*4] 4. An input panel of a resistance layer type comprising:

a first board having a first transparent conductive layer on one surface;

a second board having a second transparent conductive layer on one surface, said first board and said ...

- $\dots$  [\*4] first spacers have such a height smaller than that of said second spacers that an appropriate input sensitivity of said input panel can be achieved.
- [\*5] 5. A method of forming an input panel of a resistance layer type, said method comprising the steps of:
- a) forming first spacers on a first transparent conductive layer formed on first board, said first spacers being non-conductive and having a height ... PAGE 13

LEVEL 1 - 11 OF 225 PATENTS

5,665,502

<=2> GET 1st DRAWING SHEET OF 1

Sep. 9, 1997

Electrophotographic photoreceptor and method for producing the photoreceptor

INVENTOR: Ohashi, Kunio, Nara, Japan Tokuyama, Mitsuru, Nara, Japan

Kinashi, Hiroshi, Kyoto, Japan Nozomi, Mamoru, Kanagawa, Japan

Umehara, Tadashi, Niigata, Japan Asari, Toshiya, Kanagawa, Japan a developing gap holding jig, and in a region in contact with [\*8] cleaner. [\*9] 9. An electrophotographic photoreceptor as claimed in claim 1, wherein said photoconductive layer is a laminated layer type photoconductive layer comprising at least a charge generating layer and a charge transfer layer, said charge generating layer baving a thickness of from 0.1 to 2 mu m, and said charge transfer ...

LEVEL 1 - 12 OF 225 PATENTS

5,647,284

GET 1st DRAWING SHEET OF 6

Jul. 15, 1997

Method and apparatus for shipping knobbed glass cookware covers

including a plurality of integrally-formed flaps, each flap assuming an upwardly-angled position when one of said projections from a layer of such type of goods stacked immediately underneath the partition projects upward through an INVENTOR: Frysinger, Eric T., Groveport, Ohio Pirello, Joe, Reynoldsburg, Ohio

opening in the partition formed by said flap,

(c) a substantially rigid top plate for covering a top layer of said type of goods, the plate having a size and shape approximately equal to that of said pallet and including a plurality of equally-spaced holes to accommodate said projections from said top layer of such type of goods when said goods are stacked on a partition there beneath, said top plate further including substantially square notches inwardly-formed into its corners to complimentarily engage vertical corner posts on said pallet so ...

5,642,188

GET 1st DRAWING SHEET OF 2

Jun. 24, 1997

Wet-type electrophotographic image formation method

INVENTOR: Mochizuki, Manabu, Yokohama, Japan Kurotori, Tsuneo, Tokyo, Japan Ariyama, Kenzo, Yokohama, Japan Kojima, Kenji, Tokyo, Japan

PAGE

- ... [\*1] said silicone oil, and said photoconductive member is an organic photoconductive member.
- $[\star2]$  2. The wet-type image formation apparatus as claimed in claim 1, wherein said organic photoconductor is of a single layer type in which a charge generating material and a charge transporting material are contained.
- [\*3] 3. The wet-type image formation apparatus as claimed in claim wherein said photoconductive layer comprises ( ...
- said silicone oil, and said photoconductive member is an organic photoconductive member. [49]
- $[*10]^{\rm sol}$  10. The wet-type image formation apparatus as claimed in claim 9, wherein said organic photoconductor is of a single layer type in which a charge generating material and a charge transporting material are contained.
- [\*11] 11. The wet-type image formation apparatus as claimed in claim 9, wherein said photoconductive layer comprises ( ... LEVEL 1 - 14 OF 225 PATENTS

16

5,636,997

GET 1st DRAWING SHEET OF

Jun. 3, 1997

Protective circuit for semiconductor power device

INVENTOR: Palara, Sergio, Catania, Italy Sueri, Stefano, Catania, Italy

- ... [\*1] second circuit means comprise a condenser in an N + /P junction located in an epitaxial region contained within an insulation well of the type P in turn contained in an epitaxial layer of the type N grown on a substrate of the type N +, and an NPN transistor having as the collector an enriched region of the type n + of said epitaxial region, as the base said insulation well of the type P and as the ...
- ... [\*7] second circuit means comprise a condenser in an N + /P junction located in an epitaxial region contained within an insulation well of the type P in turn contained in an epitaxial layer of the type N-grown on a substrate of the type N + , and an NPN transistor having as the collector an enriched region of the type n + of said epitaxial region, as the base said insulation well of the type P and as the ... LEVEL 1 - 15 OF 225 PATENTS

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#. -- -- Apr. 22, 1997

Multi-layer type light emitting device

INVENTOR: Suehiro, Yoshinobu, Gyoda, Japan Yamazaki, Shigeru, Gyoda, Japan Sato, Takashi, Gyoda, Japan

What is claimed is:

[\*1] 1. A multi-layer type light emitting device, comprising:

at least two light emitting sources, successively disposed along a light-transmitting path, including a rearmost source at a rear end of the path and a ...

- ... [\*1] direction rays of light emitted by each source except the rearmost source, and for transmitting therethrough in the forward direction rays of light emitted by the rearmost source.
- [\*2] 2. A multi-layer type light emitting device according to claim 1, which further comprises a reflection surface disposed so as to correspond to said rearmost light emitting source, for reflecting rays of light emitted by said rearmost light emitting source and radiating the rays of light in the forward direction.
- $\lceil \star 3 \rceil$  3. A multi-layer type light emitting device according to claim 1, wherein said light emitting sources emit rays of light with different luminous wavelength ranges, respectively.
- [\*4] 4. A multi-layer type light emitting device according to claim 3, wherein said light emitting sources include ones emitting rays of red or nearly red light, green or nearly green light and blue or nearly blue light respectively.
- [\*5] 5. A multi-layer type light emitting device according to claim 1, wherein at least one of said light emitting sources emits ray of light with two or more luminous wavelength ranges.
- [\*6] 6. A multi-layer type light emitting device according to claim 1, wherein said at least two light emitting sources includes three light emitting sources, and wherein the respective optical surface for each light emitting source except the rearmost source each reflects rays of light emitted by said light emitting source with approximately the same luminous intensity distribution characteristics.
- [\*7] 7. A multi-layer type light emitting device according to claim 1, wherein said optical surface is a light semi-transmissible thin film reflection surface, or said optical surface is formed by a method wherein reflection

portions are partially formed on a light transmissible surface.

Pat. No. 5623181, \*7

- 8. A multi-layer type light emitting device according to claim 1, wherein said optical surface is a wavelength selective surface.
- [\*9] 9. A multi-layer type light emitting device according to claim 8, wherein said optical surface is a dichroic mirror formed by multi-layering of thin films having different refractive indexes.
- [\*10] 10. A multi-layer ...
- ... [\*10] each of said optical surface and said reflection surface of each of said sources is a concave surface facing a luminous surface of said at least one LED chip. ... [\*10]
- [\*11] 11. A multi-layer type light emitting device according to claim 10, wherein the respective optical surface and said LED chip of each source, except the rearmost source, and said reflection surface and said LED chip are each integrally formed of a first light transmissible material, thereby forming respective LEDs.
- [\*12] 12. A multi-layer type light emitting device according to claim 11, wherein each LED is disposed in such a manner that a front surface thereof is complementary to and closely connected with a rear surface of an LED of a source positioned forward thereof.
- [\*13] 13. A multi-layer type emitting device according to claim 11, further comprising a second light transmissible material having a refractive index approximately the same as that of the first light transmissible material filling
- surface or said reflection surface for the source having the LED ... [\*15] chip, and

wherein said optical surface is formed on a front surface of the light transmissible material sealing the LED of the rearmost source.

- [\*16] 16. A multi-layer type light emitting device according to claim 11, wherein an incident surface is provided on the path rearward of each optical surface, in spaced relation thereto, further comprising a further light transmissible material filling ...
- ... [\*16] optical surface, wherein rays of light emitted on the path rearward of the incident surface in the forward direction pass through the incident surface, the further light transmissible material and the optical ... [\*16]
- [\*17] 17. A multi-layer type light emitting device according to claim 1, wherein the at least two light emitting sources includes at least three light

emitting sources, successively disposed along the light-transmitting path, the at least three ...

... [\*17] source, each optical surface transmitting therethrough in the forward direction rays of light emitted by each rear source that is disposed between the optical surface and the rear end. Pat. No. 5623181, \*17

[\*18] 18. A multi-layer type light emitting device, comprising:

at least two light emitting sources, successively disposed along a light-transmitting path, including a rearmost source at a rear end of the path

transmitting therethrough in the forward direction rays of light emitted wonthe rearmost source; ... [\*18]

wherein each of the light emitting sources, except the rearmost source, is semiconductor LED chip. [\*19] 19. A multi-layer type light emitting device according to claim 18, wherein the at least two light emitting sources includes at least three light emitting sources, successively disposed along the light-transmitting path, the at least three ...

... [\*19] source, each optical surface transmitting therethrough in the forward direction rays of light emitted by each rear source that is disposed between the optical surface and the rear end.

[\*20] 20. A multi-layer type light emitting device, comprising:

at least two light emitting sources, successively disposed along a light-transmitting path, including a rearmost source at a rear end of the path and a ... ... [\*20] flat optical board formed of a light transmissible material, the light transmissible material having an annular optical surface surrounding a side surface of the at least one LED chip.

[\*21] 21. A multi-layer type light emitting device according to claim 20, wherein the at least two light emitting sources includes at least three light emitting sources, successively disposed along the light-transmitting path, the

PAGE 20

LEVEL 1 - 16 OF 225 PATENTS

5,605,051

<=2> GET 1st DRAWING SHEET OF 83

Feb. 25, 199

Automotive air conditioner having condenser and evaporator provided within air duct

INVENTOR: Iritani, Kunio, Anjo, Japan Numazawa, Shigeo, Nagoya, Japan Fujiwara, Kenichi, Kariya, Japan Yamanaka, Yasushi, Nakashima-gun, Japan Isaji, Akira, Nishio, Japan Suzuki, Takahisa, Kariya, Japan Sanada, Ryoichi, Kariya, Japan

converting a variation of temperature of the refrigerant flowing from said condenser into a variation of pressure. ... [\*13]

[\*14] 14. An automotive air conditioner according to claim 13, wherein said condenser and said subcooler are formed as one layer type heat exchanger having a large number of tubes serving as refrigerant passageway, a large number of heat radiating fins layered alternately with the tubes, and a pair of headers disposed on the opposite ends of the tubes. [\*15] 15. An automotive air conditioner according to claim 14, wherein said layer type heat exchanger comprises a partition plates in said headers in order that refrigerant flow is turned back and a mounting pipe for mounting said temperature sensitive tube.

[\*16]

LEVEL 1 - 17 OF 225 PATENTS

5,589,960

GET 1st DRAWING SHEET OF

4

Dec. 31, 1996

Liquid crystal display system

INVENTOR: Chiba, Masao, Saitama, Japan Ishii, Mikio, Saitama, Japan

What is claimed is:

[\*1] 1. A double-layer type super-twisted nematic liquid crystal display system comprising: a dot-matrix type liquid crystal display device for displaying at least one of characters and graphic forms;

a compensating ...

PAGE

- ... [\*1] crystal display device and said compensating liquid crystal device are driven with said drive voltages which are adjusted according to the calculation of contrast made with the aid of said light detecting means.
- [\*2] 2. A double-layer type super twisted nematic liquid crystal display system according to claim 1, further comprising:

a voltage memory circuit for storing most recent values of said drive voltages while said system is an off state to provide initial drive voltage values for when said system is switched to an on state.

- [\*3] 3. A double-layer type super-twisted nematic liquid crystal display system\_comprising:
- a dot-matrix type liquid crystal display device for displaying at least one of characters and graphic forms;
- a compensating ...
- ... [\*3] said light detecting means and said liquid crystal display has luminance measuring region which is turned on and off for measurement of the contrast of said liquid crystal display device.
- [\*4] 4. A double layer type super-twisted nematic liquid crystal display system comprising:

a dot-matrix type liquid crystal display device for displaying at least one of characters and graphic forms;

a compansating ...

... [\*4] liquid crystal display device has a luminance measuring region which is divided into two parts which are alternately turned on and off for measurement of the contrast of said liquid crystal display device.

Pat. No. 5589960, \*4

- [\*5] 5. A double layer type super-twisted nematic liquid crystal display system comprising:
- a dot-matrix type liquid crystal display device for displaying at least one of characters and graphic forms;

a compensating ...

LEVEL 1 - 18 OF 225 PATENTS

5,580,816

<=2> GET 1st DRAWING SHEET OF 5

Dec. 3, 1996

PAGE 2

INVENTOR: Hemmenway, Donald F., Melbourne, Florida Pearce, Lawrence G., Palm Bay, Florida

another; ... [\*1] providing an implant of a dopant species in a defined field region adjacent two of the device regions, said implant of sufficient energy and concentration to impart, nucleation sites within the device layer of the type known to result in stacking faults during oxide growth conditions;

providing a thickness of thermally grown silicon dioxide in the field regions by thermally processing the structure to remove nucleation ... 24

LEVEL 1 - 19 OF 225 PATENTS

5,575,418

GET 1st DRAWING SHEET OF 12

Nov. 19, 1996

Corrugated paperboard package systems with gas-permeable plastic membranes for modified atmosphere packaging of fresh fruits and vegetables and cut flowers

INVENTOR: Wu, Chiu H., Vancouver, Canada Oikarinen, Juhani I., Lahti, Finland Matstoms, Bo, Orebro, Sweden Powrie, William D., North Vancouver, Canada

paperboard combination so that no natural pinholes are formed. [6\*] ::  $[*10] \quad 10.$  A paperboard as claimed in claim 1 wherein the overall permeability of the paperboard combination is regulated in part by regulating the the composition of the polymer layer and the type of kraft paper.

[\*11] 11. A paperboard as claimed in claim 1 wherein the polymer is selected from the group consisting of ethylene vinylacetate (EVA), ethylbutyl acetate (EBA), a crosslinked ionomer resin, cast ... LEVEL 1 - 20 OF 225 PATENTS

5,570,084

GET 1st DRAWING SHEET OF 7

Oct. 29, 1996

Method of loose source routing over disparate network types in a packet communication network

INVENTOR: Ritter, Michael W., Los Altos, California Bettendorff, John, San Jose, California Flammer, III, George H., Cupertino, California Galloway, Brett D., Campbell, California

What is claimed is:

- [\*1] 1. A method for digital packet communication between nodes in disparate networks including path unaware network layer types and path aware network layer types, said method comprising:
- a) receiving a typed encapsulating packet which encapsulates a path-addressed packet at a first network layer, said first network layer being path aware;

... [\*1] path aware protocol if said second network layer is path aware.

[\*2] 2. A method for digital packet communication between nodes in disparate networks including path unaware network layer types and path aware network layer types, said method comprising:

a) receiving an encapsulating packet which encapsulates a path-addressed packet at a first network layer of a first type;

b) if said first type is path unaware, ...

... [\*6] type of the received packet; and

relaying the received packet to an appropriate network router.

[\*7] 7. A method for digital packet communication between nodes in various networks including path aware network layer types, said method comprising:

a) designating a destination path element for a packet by means of a type-length-value element specific only to one station of a group of ...

PAGE

LEVEL 1 - 21 OF 225 PATENTS

5.555.34

<=2> GET 1st DRAWING SHEET OF 24

Sep. 10, 1996

Method and apparatus for controlling a robot using a neural network

INVENTOR: Yoneda, Takao, Nagoya, Japan Komura, Katsuhiro, Takahama, Japan ... [\*2] accordance with the second joint angle vector calculated by said second calculation means.

[\*3] 3. An apparatus for controlling an articulated robot according to claim 2, wherein said neural network is of a three layer type which is composed of an input layer, an intermediate layer and an output layer.

[\*4] 4. An apparatus for controlling an articulated robot according to claim 2, further comprising:

actual position measurement means for measuring the actual  $\dots$ 띯

LEVEL 1 - 22 OF 225 PATENTS

5,545,945

GET 1st DRAWING SHEET OF 1

Aug. 13, 1996

Thermionic cathode

INVENTOR: Branovich, Louis E., Howell, New Jersey Eckart, Donald W., Wall, New Jersey Fischer, Paul, Oakhurst, New Jersey

... [\*赵郎 emissions.

 $[\star 3]$  3. A thermionic cathode as recited in claim 2 wherein the cathode is an impregnant-type cathode.

[\*4] 4. A thermionic cathode as recited in claim 2 wherein the cathode is a layer-type cathode.

5. An enhanced electron emission thermionic cathode, comprising: [\*5] a base material having a composition including Barium and Tungsten; and

an overcoating of emissive material forming an emissive surface on said base material: PAGE

28

LEVEL 1 - 23 OF 225 PATENTS

5,525,541

GET 1st DRAWING SHEET OF 6

Jun. 11, 1996

Method of making an electronic and/or photonic component

INVENTOR: Krauz, Philippe, Creteil, France Rao, Elchuri K., Issy-Les-Moulineaux, France

... [\*1] region of the quantum well layer on which the dielectric layer is deposited to confer on said region electro-optical or photonic properties that correspond to said function.

 $[\star 2]$  2. A method according to claim 1, wherein the quantum well layer is of the type based on GaAs and on InP.

[\*3] 3. A method according to claim 2, wherein the quantum well layer is of one of the following types: GaAs/GaAlAs; InGaAs/Ga(Al)As; InGaAs/InAlAs; InGaAs/InP; and ...

29

LEVEL 1 - 24 OF 225 PATENTS

5,504,558

<=2> GET 1st DRAWING SHEET OF

Apr. 2, 1996

Electrophotographic photosensitive member, and electrophotographic apparatus and device unit employing the

INVENTOR: Ikezue, Tatsuya, Yokohama, Japan

... [\*11] photosensitive member according to claim 10, wherein the charge-transporting layer has a thickness of from 15 to 30 mu m.

[\*12] 12. An electrophotographic photosensitive member according to claim 1, wherein the photosensitive layer is of a single layer type.

 $[\star 13]$  13. An electrophotographic photosensitive member according to claim 12, wherein the photosensitive layer has a thickness of from 10 to 35 mu m.

[\*14] 14. An electrophotographic photosensitive member according to claim 13, wherein the photosensitive layer has a ... LEVEL 1 - 25 OF 225 PATENTS

5,489,372

Feb. 6, 1996

Process for producing light absorption layer of solar cell

INVENTOR: Hirano, Tomio, Susono, Japan

39 PAGE ... [\*7] indium plating bath with a dispersion of fine particles of selenium suspended therein to form a multi-layer electrodeposited layer including copper, indium, and selenium on said conductive substrate; and

heat-treating the multi-layer type electrodeposition layer to convert it into a ternary alloy layer of copper-indium-selenium.

[\*8] 8. The process according to claim 7, wherein said copper plating bath is a sulfuric acid type electrodeposition bath which ... LEVEL 1 - 26 OF 225 PATENTS

5,475,700

GET 1st DRAWING SHEET OF 5 <=5>

Dec. 12, 1995

Laser diode with electron and hole confinement and barrier layers

INVENTOR: Iwata, Hiroshi, Tokyo, Japan

... [\*11] indices of said hole confinement layer and said electron confinement layer.

 $\lceil \star 12 \rceil - 12.$  A laser diode as claimed in claim 11, wherein said hole confinement layer and said electron confinement layer are type II compound layers. [\*13] 13. A laser diode as claimed in claim 11, wherein said hole confinement layer and said electron confinement layer are type II superlattice layers. [\*14]: 14. A laser diode as claimed in claim 11, wherein said hole potential in said hole confinement layer increases with distance away from said interface with ...

32

LEVEL 1 - 27 OF 225 PATENTS

5,466,892

m GET 1st DRAWING SHEET OF

Nov. 14, 1995

Circuit boards including capacitive coupling for signal transmission and methods of use and manufacture

INVENTOR: Howard, James R., Santa Clara, California Lucas, Gregory L., Newark, California

- ... [\*3] between sets of the additional signal and receptor pads on the first and second conductive layers also for electrostatic transmission of AC signals therebetween.
- multi-layer type having at least a third conductive layer and further wherein the additional conductive circuit means on the adjacent pairs of the first, second and third conductive layers form additional signal and receptor pads 4. The circuit board of claim 1 wherein the circuit board is of a separated by additional ...
- ... [\*9] coupling between sets of the additional signal and receptor pads on the first and second conductive layers also for electrostatic transmission of AC signals therebetween.
- $[\star 10]$  10. The method of claim 7 wherein the circuit board is of a multi-layer type having at least a third conductive layer and further wherein the additional conductive circuit means on the adjacent pairs of the first, second and third conductive layers form additional signal and receptor pads separated by additional ...
- ... [\*13] in the AC signal transmitting circuit whereby capacitive reactance and inductive reactance approach equality in order to optimize in the AC signal transmitting circuit whereby capacitive capacitive coupling.
- [\*14] 14. The circuit board of claim 13 wherein the circuit board is of a multi-layer type having at least a third conductive layer and further wherein the additional conductive circuit means on the adjacent pairs of the first, second and third conductive layers form additional signal and receptor means separated by additional ...

LEVEL 1 - 28 OF 225 PATENTS

5,466,609

<=2> GET 1st DRAWING SHEET OF 5

Nov. 14, 1995

Biodegradable gelatin-aminodextran particle coatings of and processes for making same

INVENTOR: Siiman, Olavi, Davie, Florida Burshteyn, Alexander, Hialeah, Florida Gupta, Ravinder K., Pembroke Pines, Florida CORE TERMS: gelatin, magnetic, particle, antibody, aminodextran, cell, bead, sample, ferrite, coating, minute, suspension, depletion, coated, tube, biological, rbc, crosslinked, sulfhydryl, mixed, conjugated, aqueous, layer, crosslinking, granulocyte, maleimidyl, preparation, wbc, dextran, manganese

PAGE 33

We claim:

- [\*1] 1. Colloidal particles having a plurality of pendent functional groups on an exterior coating of aminodextran in which each particle comprises a solid metallic core coated with a first gelatin layer of type B, alkali cured gelatin of Bloom in the range 60 to 225 and a second layer of an aminodextran, said layers having been either (a) crosslinked by the action of a chemical crosslinking agent or (b) ...
- pendent functional groups on an exterior coating of aminodextran in which each particle comprises a solid metallic core coated either with biodegrable, crosslinked or condensed layers of type B, alkali cured gelatin of Bloom 60 to 225 and an aminodextran, said process comprising;
- (a) (i) (1) preparing metallic core particles in said gelatin or (2) adsorbing as a  $\dots$

34

LEVEL 1 - 29 OF 225 PATENTS

5,465,103

<=2> GET 1st DRAWING SHEET OF 6

Nov. 7, 1995

Display device with coordinate input function

INVENTOR: Yoshioka, Kazuo, Nagasaki, Japan

for both displaying images and inputting coordinates, comprising:  $\cdots$  [\*1] a sensor means for sensing coordinate input detection signals from a control means:

a liquid crystal display panel of two-layer type including an optical phase compensation cell, as a first layer of the liquid crystal display panel and a liquid crystal display cell as a second layer of the liquid crystal display ...

PAGE

LEVEL 1 - 30 OF 225 PATENTS

5,441,516

വ GET 1st DRAWING SHEET OF <=5>

Aug. 15, 1995

**Temporary stent** 

INVENTOR: Wang, Lixiao, Maple Grove, Minnesota Willard, Martin R., Maple Grove, Minnesota Tran, Thomas T., Coon kapids, Minnesota Hastings, Roger, Maple Grove, Minnesota

Schmaltz, Dale F., Boulder, Colorado Holman, Thomas J., Minneapolis, Minnesota ... [\*15] closely wound helices each comprises at least two oppositely wound windings forming multiple layers.

[\*16] 16. The apparatus of claim 1 wherein the closely wound helices are each of at least the two filar double layer type.

 $[*17] \ 17.$  The apparatus of claim 16 wherein the closely wound helices are of at least the four filar double layer type.

[\*18] 18. The apparatus of claim 1 wherein a proximal elongate portion of the outer tubular member is comprised of a polymeric/braided composite joined to a distal wire wound portion.

[\*19] 19. The apparatus of claim 18 wherein the polymeric portion of the composite is polyimide.

at [\*20] 20. The apparatus of claim 18 wherein the wire wound portion is of least the four filar double layer type. [\*21] 21. The apparatus of claim 1 wherein a proximal elongate portion of the inner tubular member is comprised of a polymeric/braided composite joined to a distal wire wound portion.

 $[\star 22]$  22. The apparatus of claim 21 wherein the polymeric portion of the composite is polyimide.

[\*23] 23. The apparatus of claim 21 wherein the wire wound portion is of at least the four filar double layer type.

[\*24] 24. The apparatus of claim 1 including means connected to the proximal end portion of the catheter for introducing infusion fluid through one of the tubular members.

[\*25] 25. The apparatus of claim 24 wherein the infusion fluid  $\dots$  PAGE 36

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LEVEL 1 - 31 OF 225 PATENTS

5,432,038

<=2> GET 1st DRAWING SHEET OF 1

Jul. 11, 1995

Process for producing an organic photosensitive material preventing blushing

INVENTOR: Katsukawa, Masato, Osaka, Japan Tanaka, Masashi, Osaka, Japan ... [\*2] process for production according to claim 1, wherein the coefficient k is a number which is 0.13 or smaller.

ø [\*3] 3. A process for production according to claim 1, wherein said electrophotographic photosensitive material is of the single layer type containing a charge-generating material, a charge-transporting material and binder resin. [\*4] 4. A process for production according to claim 1, wherein said weight percentage (C) is from  $0.1\ \text{to}$  ... LEVEL 1 - 32 OF 225 PATENTS

37

5,428,244

<=2> GET 1st DRAWING SHEET OF 12

Jun. 27, 1995

Semiconductor device having a silicon rich dielectric layer

INVENTOR: Segawa, Mizuki, Kyoto, Japan Kato, Yoshiaki, Hyogo, Japan Nakaoka, Hiroaki, Osaka, Japan

comprising a dielectric layer for passivation, said passivation dielectric layer being formed on said dielectric layer type, being composed of a chemical compound which is the same compound that said dielectric layer is composed of a chemical and having a silicon content closer to a silicon content according to the stoichiometric composition formula, compared to the dielectric layer.

LEVEL 1 - 33 OF 225 PATENTS

5,420,052

GET 1st DRAWING SHEET OF

May 30, 1995

Method of fabricating a semiplanar heterojunction bipolar

transistor

INVENTOR: Morris, Francis J., Plano, Texas Yang, Jau-Yuann, Richardson, Texas Plumton, Donald L., Dallas, Texas Yuan, Han-Tzong, Dallas, Texas

... [\*1] layer;

forming a collector plug region through a selected portion of the collector layer to the subcollector layer:

forming a base layer on the collector layer and the collector plug region;

forming an emitter layer type on the base layer;

forming an emitter cap layer on the emitter layer;

forming a collector contact on the collector plug region;

forming an emitter contact on the emitter cap layer;

forming a base contact on the ...
LEVEL 1 - 34 OF 225 PATENTS

5,401,549

Mar. 28, 1995

Optical information recording medium

INVENTOR: Watase, Kenta, Tokyo, Japan

... [\*1] layer is overlaid on both said reflective layer in said ROM region portion and a recording portion of the substrate in said recording region, said recording layer being of a multi-layer type comprising a dielectric layer and a magnetic layer.

[\*2] 2. The optical information recording medium as claimed in claim 1, further comprising an additional reflective layer which is overlaid on said recording ...

LEVEL 1 - 35 OF 225 PATENTS

5,390,208

<=2> GET 1st DRAWING SHEET OF 3

Feb. 14, 1995

Strained quantum well type semiconductor laser device

INVENTOR: Kasukawa, Akihiko, Tokyo, Japan

Kikuta, Toshio, Tokyo, Japan

What is claimed is:

[\*1] 1. A strained quantum well layer type semiconductor laser device comprising a light emitting active layer of a multilayer structure including a quantum well layer and a barrier layer and a pair of light confining layers ...

... [\*1] y P 1 - y ( $\theta$  < y </= 1) and the barrier layer and/or the light confining a yers are made of In 1 - x Ga x P ( $\theta$  < x </= 1).

SYMBOL OMITTED epsilon w x L w SYMBOL OMITTED < 45(% x nm),

where w is the ratio of deformation (%) and Lw is the thickness (nm) of each component layer of the strained quantum well layer.

[\*3] 3. A strained quantum well layer type semiconductor laser device according to claim 1, wherein the In 1-x Ga x P barrier layer and each of the In 1-x Ga x P light confining layers satisfy ...

... [\*3] OMITTED < 45(% x nm),

where s is the ratio of deformation (%) and Ls is the thickness (nm) of each component layer of the barrier layer and the light confining layers.

[\*4] 4. A strained quantum well layer type semiconductor laser device according to claim 1, wherein InAs y P 1 - y (0 < y </= 1) has a value for compositional ratio y between 0.3 and 0.6. LEVEL 1 - 36 OF 225 PATENTS

5,387,564

Feb. 7, 1995

Molding and calcining of zeolite powder

INVENTOR: Takeuchi, Tatsuro, Tsukuba, Japan Mouri, Motoya, Tsuchiura, Japan Okabayashi, Saji, Kitakanbara, Japan Miyamura, Shoichi, Kitakanbara, Japan

... [\*21] a) a zeolite;

- (b) a teta 1,3-glucan in an amount of 0.1-20 parts by weight in relation to 100 parts by weight of the zeolite; and
- (c) a 1:1 layer-type clay mineral and a 2:1 layer clay mineral in a total amount of 5-50 parts by weight in relation to 100 parts by weight of the zeolite; and

(ii) at least ...

LEVEL 1 - 37 OF 225 PATENTS

5,374,328

σ GET 1st DRAWING SHEET OF

Dec. 20, 1994

PAGE

# Method of fabricating group III-V compound

INVENTOR: Remba, Ronald D., Sunnyvale, California Brunemeier, Paul E., Sunnyvale, California Schmukler, Bruce C., Mountain View, California Strifler, Walter A., Sunnyvale, California Rosenblatt, Daniel H., Belmont, California

... [\*5] 1-x As wherein (0 </= y < 0.2) and (0.2 < x </= 1.0)

6. A method of making a semiconductor device comprising the steps of: [9\*]

fabricating a structure by

(i) growing one or more layers of the type X a Y 1-a As, where X is an atom selected from the group of IIIA atoms and Y is a different atom selected from the group of IIIA atoms, and where (0 < a </= 1) upon a semiconductor ... PAGE 43

LEVEL 1 - 38 OF 225 PATENTS

to the same

5,324,980

<=2> GET 1st DRAWING SHEET OF 24

Jun. 28, 1994

Multi-layer type semiconductor device with semiconductor element layers stacked in opposite direction and manufacturing method thereof

INVENTOR: Kusunoki, Shigeru, Hyogo, Japan

What is claimed is:

[\*1] 1. A multi-layer type semiconductor device, comprising:

a substrate having a main surface;

a first semiconductor element layer formed on said main surface of said substrate and including a first semiconductor element having an active ...

... [\*3] insulating layer.

[\*4] 4. The device of claim 3, including a conductor filling said through-hole and contacting opposite surfaces of said first and second semiconductor element layers.

[\*5] 5. A multi-layer type semiconductor device, comprising:

a substrate having a main surface;

a first semiconductor element layer formed on said main surface of said substrate and including a first semiconductor element having an active ...

... [\*5] contact with said insulating layer and being oriented back-to-back on said first and second semiconductor element layers, each of said regions including one or more of the semiconductor elements.

[\*6] [\*6] [\*6]. A multi-layer type semiconductor device, comprising:

a substrate having a main surface;

a first semiconductor element layer formed on said main surface of said substrate and including a first semiconductor element having an active ...

... [\*6] second semiconductor element layers.

[\*7] 7. The device of claim 6, wherein a through-hole is formed only through said field oxide layer and said interlayer insulation film.

[\*8] 8. A multi-layer type semiconductor device, comprising:

a substrate having a main surface;

Pat. No. 5324980, \*8

a first semiconductor element layer formed on said main surface of said substrate and including a first semiconductor element having an active ... PAGE 45

LEVEL 1 - 39 OF 225 PATENTS

5,324,678

<=2> GET 1st DRAWING SHEET OF 24

Jun. 28, 1994

Method of forming a multi-layer type semiconductor device with semiconductor element layers stacked in opposite

directions

INVENTOR: Kusunoki, Shigeru, Hyogo, Japan

What is claimed is:

 $[\star 1]$  1. A method of manufacturing a multi-layer type semiconductor device comprising the steps of;

forming a base member by successively stacking, on a main surface of a first substrate, a first semiconductor layer, an insulating layer and a second ...

 $\dots$  [\*1] semiconductor device by using said first semiconductor layer as a base with an exposed surface of said first semiconductor layer directed upward.

[\*2] 2. A method of manufacturing a multi-layer type semiconductor device according to claim 1, wherein the step of forming the base member includes the steps of;

bonding a first member including said first semiconductor layer formed on said first substrate, with a ...

... [ $\star 2$ ] said insulating layer and said first semiconductor layer are opposed to each other, and

thinning said third substrate the expose said second semiconductor layer.

[\*3] 3. A method of manufacturing a multi-layer type semiconductor device comprising the steps of;

forming perforations through a first substrate;

filling said perforations with conductors

successively forming a first semiconductor layer on a main surface of

LEVEL 1 - 40 OF 225 PATENTS

œ GET 1St DRAWING SHEET OF

Mar. 1, 1994

LED carriage selectively movable in two directions

INVENTOR: Isobe, Minoru, Tokyo, Japan

end fixed to one end of the first block, and the other end fixed to a right end of the frame, a stacked-layer type of second piezoelectric element adapted to deform itself in response to an application of a voltage by a power source for deforming the second leaf spring, the second piezoelectric element being fixed

... [\*5] comprises an L-shaped first block, an L-shaped leaf spring having one end fixed to one end of the first block, and the other end fixed to one end of a frame, a stacked-layer type of second piezoelectric element adapted to deform itself in response to an application of a voltage by a power source for deforming the leaf spring, the second piezoelectric element being fixed to the

end fixed to one end of the first block, an L-shaped second leaf spring having one of fixed to one end of the first block, and the other end fixed to a right end of the frame, a stacked-layer type of second piezoelectric element adapted to deform itself in response to an application of a voltage by a power source for

deforming the second leaf spring, the second piezoelectríc element being fixed

PAGE ... [\*20] comprises an L-shaped first block, an L-shaped leaf spring having one end fixed to one end of the first block, and the other end fixed to one end of a frame, a stacked-layer type of second piezoelectric element adapted to deform itself in response to an application of a voltage by a power source for deforming the leaf spring, the second piezoelectric element being fixed to the

LEVEL 1 - 41 OF 225 PATENTS

5,289,486

S GET 1st DRAWING SHEET OF

Feb. 22, 1994

 $_{\mathrm{SPR}_{\mathrm{sp}}}$  Semiconductor luminous element and superlattice structure

INVENTOR: Iga, Kenichi, Machida, Japan Koyama, Fumio, Hino, Japan Takagi, Takeshi, Ibaraki, Japan ... [\*8] between the active layer and the multi-quantum barrier layer.

[\*9] 9. A superlattice structure, comprising alternating layers of at least two types of crystals having different energy gaps,

wherein the energy gaps of adjacent crystal layers are such that the type of crystal having the smaller energy gap of the adjacent layers has an energy gap which is smaller than that of a portion of the superlattice structure on a side through which electrons or holes enter the superlattice structure, and

wherein the thicknesses and structures of the adjacent crystal ...

LEVEL 1 - 42 OF 225 PATENTS

GET 1st DRAWING SHEET OF

Jan. 4, 1994

Method of producing an absorber layer for solar cells with the aid of electrodeposition

INVENTOR: Bonnet, Dieter, Friedrichsdorf, Federal Republic of Germany Ehrhardt, Josef, Hochheim/Main, Federal Republic of Germany Hewig, Gert, Alzenau, Federal Republic of Germany

... [\*1] electroplating bath and simultaneously incorporating a third alloy component of Group VIA suspended in the electroplating bath in finely dispersed form by dispersion electrolysis, and

producing a ternary semiconductor layer of the type IB-IIIA-VIA by heat treating the deposited material.

[\*2] 2. The method according to claim 1, wherein the volume percentage of each component of the binary alloy is between about 25% and 75%.

3. The method ... LEVEL 1 - 43 OF 225 PATENTS [\*3]

Dec. 21, 1993

Benzidine derivative and photosensitive material using said derivative

INVENTOR: Hanatani, Yasuyuki, Osaka, Japan [wasaki, Hiroaki, Osaka, Japan

- photosensitive layer which contains the benzidine derivative (1) according to claim 1.
- [\*3] 3. The photosensitive material according to claim 2, wherein the photosensitive layer is a multi-layer type photosensitive layer comprising an electric charge transferring layer and an electric charge generating layer which are laminated mutually.
- [\*4] 4. The multi-layer type photosensitive material according to claim 3, wherein the electric charge transferring layer contains 25 to 200 parts by weight of said benzidine derivative (1) for 100 parts by weight of a ...
- compounds, and pyrrolopyrrole compounds. ... [\*5]
- 6. The photosensitive material according to claim 5, wherein the electric charge generating material is an azo compound.
- [\*7] 7. The photosensitive material according to claim 2, wherein the photosensitive layer is a single-layer type photosensitive layer comprised of electric charge transferring material, an electric charge generating material and a binding resin.
- [\*8] 8. The photosensitive material according to claim 7, wherein the single-layer type photosensitive layer contains 40 to 200 parts by weight of said benzidine derivative (1) for 100 parts by weight of a binding resin. [\*8]
- single-layer type photosensitive layer contains, for 100 parts by weight of [\*9] 9. The photosensitive material according to claim 8, wherein the

20

binding resin, 5 to 500 parts by weight of one or more kinds of an electric charge generating material selected from selenium, ...
LEVEL 1 - 44 OF 225 PATENTS

5,260,723

<=2> GET 1st DRAWING SHEET OF 10

Nov. 9, 1993

Liquid jet recording head

INVENTOR: Naruse, Osamu, Kanagawa, Japan Ameyama, Minoru, Kanagawa, Japan Matsumoto, Syuzo, Kanagawa, Japan Komai, Hiromichi, Kanagawa, Japan Hirata, Tositaka, Tokyo, Japan ... [\*1] between 0.01 Kg/mm<2 > and 300 Kg/mm<2> .

[\*2] 2. A liquid jet recording head as claimed in claim 1 wherein said piezo-electric member is a layer-type piezo-electric member.

[\*3] 3. A liquid jet recording head as claimed in claim 1 further comprising a driver unit driving said piezo-electric elements.

[\*4] 4. ...

... [\*6] flow paths.

[\*7] 7. A liquid jet recording head as claimed in claim 6 wherein each of said first piezo-electric member and said second piezo-electric member is a layer-type piezo-electric member.

[\*8] 8. A liquid jet recording head as claimed in claim 6 wherein said elasticity member has a modulus of elasticity between 0.01 Kg/mm<2 > and ...

... [\*13] elements in a direction perpendicular to said second flow paths.

[\*14] 14. A liquid jet recording head as claimed in claim 13 wherein said piezo-electric member is a layer-type piezo-electric member.

[\*15] 15. A liquid jet recording head as claimed in claim 13 wherein said elasticity member has a modulus of elasticity between 0.01 Kg/mm<2 > and ...

elements in the perpendicular direction to each of said plurality ... [\*18] of flow paths.

[\*19] 19. A liquid jet recording head as claimed in claim 18 wherein said piezo-electric member is a layer-type piezo-electric member.

LEVEL 1 - 45 OF 225 PATENTS

5,258,251

Nov. 2, 1993

Hydrazone compound and photosensitive material using said compound

INVENTOR: Hanatani, Yasuyuki, Sakai, Japan Iwasaki, Hiroaki, Hirakata, Japan

- same or different from one another, and each is a hydrogen atom, a halogen atom, an alkyl group or an alkoxy group.
- photosensitive layer is a multi-layer type photosensitive layer including an electric charge transferring layer which includes the hydrazone compound as the electric charge transferring material, and an electric charge generating layer, 2. The photosensitive material according to claim 1, wherein the which layers are laminated mutually.
- [\*3] 3. The photosensitive material according to claim 2, wherein the electric charge transferring layer of the multi-layer type photosensitive layer further includes a binding resin, and the electric charge transferring layer contains 25 to 200 parts by weight of said hydrazone compound for 100 parts by weight of the ...
- compounds, and pyrrolopyrrole compounds. ... [\*4]
- 5. The photosensitive material according to claim 4, wherein the electric charge generating material is an azo compound.
- 등 [\*6] 6. The photosensitive material according to claim 1, wherein the photosensitive layer is a single-layer type photosensitive layer comprised of electric charge transferring material, an electric charge generating material and a binding resin.
- [\*7] 7. The photosensitive material according to claim 6, wherein the single-layer type photosensitive layer contains 40 to 200 parts by weight of said hydrazone compound for 100 parts by weight of the binding resin.
- [\*8] 8. The photosensitive material according to claim 7, wherein the single-layer type photosensitive layer contains, for 100 parts by weight of the binding mesin, 2 to 20 parts by weight of one or more kinds of an electric charge generating material selected from selenium, selenium- ... E 52

LEVEL 1 - 46 OF 225 PATENTS

51

PAG

## Oct. 26, 1993

Hydrazone compound and photosensitive material using said compound

INVENTOR: Hanatani, Yasuyuki, Sakai, Japan Iwasaki, Hiroaki, Hirakata, Japan

- ... [\*1] halogen atom, an alkyl group, an alkoxy group or the following group: [See Original Patent for Chemical Structure Diagram].
- [\*2] 2. The photosensitive material according to claim 1, wherein the photosensitive layer is a multi-layer type photosensitive layer including an electric charge transferring layer which includes the hydrazone compound as the electric charge transferring material, and an electric charge generating layer, which layers are laminated mutually.
- [\*3] 3. The photosensitive material according to claim 2, wherein the electric charge transferring layer of the multi-layer type photosensitive layer further includes a binding resin, and the electric charge transferring layer contains 25 to 200 parts by weight of said hydrazone compound for 100 parts by weight of the ...
- ... [\*4] compounds, and pyrrolopyrrole compounds.
- 5. The photosensitive material according to claim 4, wherein the [\*5] 5. The photosensitive material according to c electric charge generating material is an azo compound.
- an [\*6] 6. The photosensitive material according to claim 1, wherein the photosensitive layer is a single-layer type photosensitive layer comprised of electric charge generating material and a binding resin.
- [\*7] 7. The photosensitive material according to claim 6, wherein the single-layer type photosensitive layer contains 40 to 200 parts by weight of said hydrazone compound for 100 parts by weight of the binding resin.
- [\*8] 8. The photosensitive material according to claim 7, wherein the single-layer type photosensitive layer contains, for 100 parts by weight of the binding resin, 2 to 20 parts by weight of one or more kinds of an electric charge generating material selected from selenium, selenium-...

LEVEL 1 - 47 OF 225 PATENTS

5,254,423

GET 1st DRAWING SHEET OF

0ct. 19, 1993

Electrophotographic photosensitive member, and electrophotographic apparatus, device unit and facsimile machine having the photosensitive member

INVENTOR: Mayama, Shinya, Yamato, Japan Fujimura, Naoto, Yokohama, Japan Yoshihara, Toshiyuki, Inagi, Japan Sakai, Kiyoshi, Hachioji, Japan Anayama, Hideki, Yokohama, Japan Ainoya, Hideyuki, Tokyo, Japan Aoki, Katsumi, Yokohama, Japan ... [\*21] 21. The electrophotographic photosensitive member according to claim 19, wherein said charge generation layer is the surface layer.

[\*22]  $_{\rm H}$  22. The electrophotographic photosensitive member according to claim 18, wherein said photosensitive layer is of a single layer type.

[\*23] 23. The electrophotographic photosensitive member according to claim 1, wherein said surface layer is a surface protective layer.

[\*24] 24. The electrophotographic photosensitive member according to claim 1, wherein said electrophotographic photosensitive member has ... GE 54

LEVEL 1 - 48 OF 225 PATENTS

5,247,445

<=2> GET 1st DRAWING SHEET OF 5

Sep. 21, 1993

Control unit of an internal combustion engine control unit utilizing a neural network to reduce deviations between exhaust gas constituents and predetermined values

INVENTOR: Miyano, Hideyo, Niza, Japan Suzaki, Yukihiko, Nerima, Japan Takahashi, Fumitaka, Hoya, Japan Ogasawara, Ken-ichi, Fujimi, Japan ... [\*7] units as the number of cylinders, and an intermediate layer arranged between said input layer and said output layer; and wherein the units are coupled with predetermined coupling weights only across the layers to form a three-layer type perceptron neural network.

[\*8] 8. A control unit for an internal combustion engine according to claim 7 wherein said control means corrects said coupling weights among the units by applying a back propagation learning method to said three-layer type perceptron neural network, and corrects the correction coefficient for said calculation means, [\*9] 9. A control unit for an internal combustion engine according to claim 7 wherein said control means corrects ...

arranged between said input layer and said output layer; and wherein the units are coupled with predetermined coupling weights only across the layers to form a three-layer type perceptron neural network.

[\*23] 23. A control unit for an internal combustion engine according to claim 22 wherein said control means corrects said coupling weights among the units by applying a back propagation learning method to said three-layer type perceptron neural network, and corrects the correction coefficient for said

[\*24] 24. A control unit for an internal combustion engine according to claim 22 wherein said control means corrects ... LEVEL 1 - 49 OF 225 PATENTS

5,244,561

<=2> GET 1st DRAWING SHEET OF 3

Sep. 14, 1993

Process and apparatus for the electrochemical determination of pCO2 in blood

INVENTOR: Calzi, Claudio, Milan, Italy
Tancredi, Gabrio, Milan, Italy

... [\*3] 3. The process of claim 1, wherein the measurement liquid is high-purity water.

 $\left[\star 4\right]$  4. The process of claim 1, wherein the measuring cell is conductivity cell.

[\*5] 5. The process of claim 4, wherein the conductivity cell is of the thin layer type.

 $[\star 6]$  6. The process of claim 1, wherein the means of removing ionic impurities are ion exchangers.

[\*7] 7. The process of 6, wherein the ion exchangers are in mixed bed form.

[\*8] [\*] 8. The process of claim 7, wherein the ion exchangers ..

... [\*12] 12. The apparatus of claim 10, wherein the measurement liquid is high-purity water.

[\*13] 13. The apparatus of claim 10, wherein the measuring cell is conductivity cell.

[\*14] 14. The apparatus of claim 13, wherein the conductivity cell is of the thin layer type.

[\*15] 15. The apparatus as claimed in claim 10, wherein the means for removing ionic impurities are ion exchangers.

 $\left[*16\right]$  16. The apparatus of claim 10, wherein the ion exchangers are in mixed bed form.

LEVEL 1 - 50 OF 225 PATENTS [\*17] 17. The ...

5,242,839

9 <=2> GET 1st DRAWING SHEET OF

Sep. 7, 1993

Method of manufacturing an integrated photoelectric

receiving device

... [\*1] layer, an etching stopper layer and an absorption layer on the substrate etched; INVENTOR: Oh, Kwang-Ryong, Daejeon, Republic of Korea Lee, Yong-Tak, Daejeon, Republic of Korea

c) removing the absorption layer excluding the photodetector forming area on the substrate by the selective etchant;

d) sequentially removing the etching stopper layer and the type n-channel layer between the photodetector and the transistor forming areas to electrically insulate the photodetector and the transistor; e) sequentially forming a p-type InP layer and a p-type InGaAs layer on the

57

LEVEL 1 - 51 OF 225 PATENTS

5,240,964

<=2> GET 1st DRAWING SHEET OF 1

Aug. 31, 1993

Process for producing urethane foam with high density skin

INVENTOR: Ohmura, Hirokazu, Niiza, Japan Yoshimura, Kimio, Urawa, Japan Narumi, Satoshi, Tochigi, Japan

What is claimed is:

28 [\*1] 1. A process for producing a urethane foam having a high density outer surface layer, of the type wherein a plastic liquid containing di or polyisocynates, polyols, a catalyst, low-molecular polyols used as a crosslinker or a chain extender, a blowing agent consisting of water, and an assistant is

LEVEL 1 - 52 OF 225 PATENTS

5,236,755

GET 1st DRAWING SHEET OF

Aug. 17, 1993

Optical recording elements

INVENTOR: Howe, Steven D., Suffolk, England Dorey, Loth Y., Essex, England

... [\*5] element as claimed in claims 1, 2, 3, or 4 in which the element is in the form of a tape.

[\*6] 6. An element as claimed in any one of claims 1 to 4, wherein the absorbing layer is of the type which is thermally deformed to form optically readable pits when subject to heating by laser radiation of said given wavelength 7. An optical recording element as claimed in any one of claims 1 to

LEVEL 1 - 53 OF 225 PATENTS

5,214,664

GET 1st DRAWING SHEET OF 15

May 25, 1993

Multiple wavelength semiconductor laser

INVENTOR: Paoli, Thomas L., Los Altos, California

emitting multiple wavelength solid state laser, comprising: ... [\*4] a plurality of contiguous layers of semiconductor material deposited on a substrate, one of said layers comprising a multiple quantum well active layer of the type wherein at least two quantum wells contained therein are formed such that one quantum level of a first quantum well is at the same energy level as a different quantum level of the second quantum well;

resonant ...

multiple wavelength solid state laser, comprising: ... [\*18]

- a substrate;
- a first cladding layer disposed on said substrate;

a multiple quantum well active layer disposed on said first cladding layer of the type having at least two adjacent quantum wells, a first of said quantum wells having a first energy bandgap between the lowest energy level of its conduction band and the uppermost level of its valence band and a ...

multiple wavelength solid state laser, comprising: ... [\*23]

a substrate

a first cladding layer disposed on said substrate;

of the type having at least two adjacent quantum wells, a first of said quantum wells having a first thickness and a second of said quantum wells having a second thickness which is greater than said first thickness, ... a multiple quantum well active layer disposed on said first cladding layer

LEVEL 1 - 54 OF 225 PATENTS

5,213,926

May 25, 1993

Phenylenediamine derivative and photosensitive material using said derivative

INVENTOR: Hanatani, Yasuyuki, Sakai, Japan Iwasaki, Hiroaki, Hirakata, Japan

photosensitive layer which containing a conductive substrate having thereon according to claim 1.

photosensitive layer is a multi-layer type photosensitive layer comprising an electric charge transferring layer on an electric charge generating layer which 3. The photosensitive material according to claim 2, wherein the are laminated mutually. [\*4] 4. The multi-layer type photosensitive material according to claim 3, wherein the electric charge transferring layer contains 25 to 200 parts by weight of said phenylenediamine derivative (1) for 100 parts by weight of a ...

compounds, and pyrrolopyrrole compounds. ... [\*5] 6. The photosensitive material according to claim 5, wherein the electric charge generating material is an azo compound.  $\lceil \star 7 \rceil$  7. The photosensitive material according to claim 2, wherein the photosensitive layer is a single-layer type photosensitive layer comprised of an

[\*8] 8. The photosensitive material according to claim 7, wherein the single-layer type photosensitive layer contains 40 to 200 parts by weight of said phenylenediamine derivative (1) for 100 parts by weight of a binding resin. electric charge transferring material, an electric charge generating material and a binding resin.

single-layer type photosensitive layer contains, for 100 parts by weight of binding resin, 2 to 20 parts by weight of one or more kinds of an electric charge generating material selected from the group ... LEVEL 1 - 55 OF 225 PATENTS 9. The photosensitive material according to claim 8, wherein the

5,200,969

GET 1st DRAWING SHEET OF 15

Apr. 6, 1993

Switchable multiple wavelength semiconductor laser

INVENTOR: Paoli, Thomas L., Los Altos, California

two different output wavelengths, comprising: .. [\*1] a laser body of the type including a plurality of contiguous layers of semiconductor material, located in an optical path, at least first and second portions of said layers of the type providing carrier quantization in at least one dimension, wherein said first and second portions are formed such that one quantum fevel of said first portion is at the same energy level as a different quantum ...

different output wavelengths, comprising: ... [\*2] a laser body of the type including a plurality of contiguous layers of semiconductor material, located in an optical path, at least a first and second portions of said layers of the type providing carrier quantization in at least one dimension, wherein said first and second portions thereof are formed such that one quantum level of said first portion is at the same energy level as a

LEVEL 1 - 56 OF 225 PATENTS

5,196,143

Mar. 23, 1993

Mixed metal hydroxide-clay adducts as thickeners for water and other hydrophylic fluids

INVENTOR: Burba, III, John L., Angleton, Texas Barnes, Audrey L., Lake Jackson, Texas

14. The adduct or reaction product of claim 1 wherein the mineral [\*14] 14. The adduct or reaction product of claim 1 wherein the mineral clay is at least one of the classes consisting of amorphous clays of the allophane group and crystalline clays of the 2-layer type, 3-layer type, expanding type, non-expanding type, elongate, regular mixed layer type, and chain structure type.

[\*15], 15. The adduct or reaction product of claim 1 wherein the mineral clay is bentonite.

[\*16] 16. The adduct or reaction product of claim 1 wherein the mineral clay is beneficiated bentonite.

17. The adduct or reaction ... LEVEL 1 - 57 OF 225 PATENTS

[\*17]

5,189,567

GET 1st DRAWING SHEET OF

Feb. 23, 1993

High speed switching circuit for controlling current flow in a bridge circuit coil for use in a magneto-optic direct overwrite system

INVENTOR: Mody, Hemant K., Rochester, New York

What is claimed is:

[\*1] 1. A direct over-write magneto-optic recording apparatus for recording digital information in a magnetic recording layer of the type having vertically oriented magnetic domains, said digital information identified by a digital information source, said apparatus comprising:

(a) means for scan-irradiating the recording layer with a beam of

by claim 2, wherein said switching elements are comprises of ... [\*3] triacs. [\*4] 4. A direct over-write magneto-optic recording apparatus for recording digital information in a magnetic recording layer of the type having vertically oriented magnetic domains, wherein an information data source provides control signals identifying data to be stored by said recording apparatus, said apparatus comprising:

(a) means for scan- ...

alternates directions during each cycle of said predetermined ... [\*6] frequency.

[+7] [4] [4] [4] [4] A direct over-write magneto-optic recording apparatus for recording digital information in a magnetic recording layer of the type having magnetic domains with a plurality of vertical orientations, wherein said digital information to be recorded is represented by control signals determining a representation of said digital information by said recording layer, ...

AGE 64

LEVEL 1 - 58 OF 225 PATENTS

5,189,500

<=2> GET 1st DRAWING SHEET OF 24

Feb. 23, 1993

Multi-layer type semiconductor device with semiconductor element layers stacked in opposite directions and manufacturing method thereof

INVENTOR: Kusunoki, Shigeru, Hyogo, Japan

What is claimed is:

[\*1] 1. A multi-layer type semiconductor device comprising;

a transparent substrate,

a photosensor layer formed on said transparent substrate and including photosensor elements for detecting light passing through said transparent substrate and converting the ...

- ... [\*1] photosensor elements of said photosensor layer and electrically connected to said circuit layer via said through holes for displaying results of processing output from said circuit layer.
- [\*2] 2. A multi-layer type semiconductor device according to claim 1, further comprising a light shielding layer interposed between said photosensor layer and said circuit layer for preventing light traveling through said photosensor layer toward said circuit layer from entering said circuit layer.
- [\*3] 3. A multi-layer type semiconductor device according to claim 1, which is formed of materials penetrable to light as a single chip.
- [\*4] 4. A multi-layer type semiconductor device according to claim 3, which, formed as the single chip, has a light transmittance of at least 5%
- [\*5] 5. A multi-layer type semiconductor device according to claim 1, wherein said display element layer includes transmission type liquid crystal display elements for giving a display based on variations of light passing therethrough, ...

- $\dots$  [\*5] layer formed between said display element layer and said circuit layer and including light emitting elements for projecting light to said liquid crystal display elements.
- [\*6] 6. A multi-layer type semiconductor device according to claim 5, further comprising a light shielding layer interposed between said light emitting element layer and said circuit layer for preventing light traveling from said light emitting element layer toward said circuit layer from entering said circuit layer.
- [\*7] 7. A multi-layer type semiconductor device according to claim 1, wherein said display element layer includes reflection type liquid crystal display elements for giving a display based on variations of reflected light.

Pat. No. 5189500, \*7

- Ø [\*8] 8. A multi-layer type semiconductor device according to claim 1, wherein said display element layer includes light emitting elements for giving display based on self-emission of light.
- [\*9] 9. A multi-layer type semiconductor device according to claim 8, further comprising a light shielding layer interposed between said display element layer and said circuit layer for preventing light traveling from said display element layer toward said circuit layer from entering said circuit
- [\*10] 10. A multi-layer type semiconductor device comprising;
- a transparent substrate,
- a display element layer including display elements and formed on said transparent substrate such that a display given by said display elements is
- ... [\*10] connected to said circuit layer via said through holes for converting an amount of information received from outside into an electric signal for processing by said circuit layer.
- [\*11] 11. A multi-layer type semiconductor device according to claim 10, wherein said sensor layer includes a photosensor element for detecting light.
- $[*12] \,$  12. A multi-layer type semiconductor device according to claim 10, wherein said sensor layer includes a temperature sensing element for detecting temperature.
- 13. A multi-layer type semiconductor device according to claim 10, wherein said sensor layer includes a pressure sensing element for detecting [\*13] pressure
- [\*14] 14. A multi-layer type semiconductor device according to claim 10, wherein said sensor layer includes a sensing element for detecting radiation.

[\*15] 15. A multi-layer type semiconductor device comprising;

a substrate defining perforations and having conductors formed in said

a first circuit layer formed on said substrate and including an electric circuit electrically connected to said ...
LEVEL 1 - 59 OF 225 PATENTS

5,189,297

<=2> GET 1st DRAWING SHEET OF 2

Feb. 23, 1993

9

Planar double-layer heterojunction HgCdTe photodiodes and methods for fabricating same

INVENTOR: Ahlgren, William L., Goleta, California

... [\*19] atoms selected for type-converting the underlying collector layer to an opposite type of electrical conductivity;

illuminating the dopant layer and the underlying surface of the collector layer with the source; and

diffusing the dopant layer into the underlying collector layer thereby type-converting the underlying collector layer to an opposite type of conductivity.

[\*20] 20. A method as defined in claim 13 wherein the step of forming base layer is accomplished by forming an n-type ...

... [\*21] depositing the liberated dopant atoms over the surface of the collector layer, the dopant atoms being deposited upon the surface only where photodiode is not desired; and

diffusing the deposited dopant atoms into the underlying collector layer thereby type-converting the underlying collector layer to an n-type of conductivity. [\*22] 22. A method as defined in claim 13 and further comprising a step of depositing a passivation layer at least over the ... LEVEL 1 - 60 OF 225 PATENTS

5,187,680

<=2> GET 1st DRAWING SHEET OF 15

Feb. 16, 1993

. 17

INVENTOR: Engeler, William E., Scotia, New York

... [\*11] lines that are identified by the same ordinal number connected to be receptive of the same input signal, thereby to provide a neural network

[\*12] 12. A plurality, L in number, of neural net layers of the type set forth in claim 11, respectively identified by consecutive ordinal numbers zeroeth through (L - 1), L being a positive integer, the non-linear amplifiers of the processors ... ... [\*18] lines that are identified by the same ordinal number connected to be receptive of the same input signal, thereby to provide a neural network

[\*19] 19. A plurality, L in number, of neural net layers of the type set forth in claim 18, respectively identified by consecutive ordinal numbers zeroeth through (L - 1), L being a positive integer, the non-linear amplifiers of the processors ... ... [\*27] lines that are identified by the same ordinal number connected to be receptive of the same input signal, thereby to provide a neural network

[\*28] 28. A plurality, L in number, of neural net layers of the type forth in claim 27, respectively identified by consecutive ordinal numbers zeroeth through (L - 1) , L being a positive integer, the non-linear amplifiers of the processors ... LEVEL 1 - 61 OF 225 PATENTS

5,185,228

Feb. 9, 1993

Electrophotosensitive material containing p-benzylbiphenyl

INVENTOR: Maeda, Tatsuo, Kobe, Japan Katsukawas Masato, Ibaraki, Japan Iwasaki, Hiroaki, Hirakata, Japan Mizuta, Yasufumi, Kishiwada, Japan ... [\*3] benzylbiphenyl is included in an amount of 20 to 150 parts by weight for 100 parts by weight of m-phenylenediamine.

[\*4] 4. An electrophotosensitive material according to claim 2, wherein the layer is a single-layer type photosensitive layer containing a charge generating

- $[\star 5]$  5. An electrophotosensitive material according to claim 4, wherein the charge generating material is a perylene compound.
- [\*6] 6. An electrophotosensitive material according to claim 2, wherein the

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contained in an amount of 20 to 150 parts by weight for 100 parts ... [\*/] Contained in an amount o' by weight of charge transfer material. [\*8] 8. An electrophotosensitive material according to claim 1, wherein the layer is a single-layer type photosensitive layer containing a charge generating material.

9. An electrophotosensitive material according to claim 8, wherein the [\*9] 9. An electrophotosensitive material acc charge generating material is a perylene compound. 10. An electrophotosensitive material according to claim 1, wherein [\*10]

LEVEL 1 - 62 OF 225 PATENTS

5,179,457

GET 1st DRAWING SHEET OF

Jan. 12, 1993

Liquid crystal display device with birefringent film between the substrates of the liquid crystal

NVENTOR: Hirataka, Jun-ichi, Hitachi, Japan

Kondo, Katsumi, Katsuta, Japan Tomioka, Yasushi, Hatoyama, Japan Imazeki, Shuji, Hatoyama, Japan Taniguchi, Yoshio, Hino, Japan

... [\*9] pair of electrode structures.

a liquid crystal layer sandwiched between said pair of substrates and electrode structures, and

having a different birefringent property and said pattern is arranged within the employed wherein a pattern is formed in said structure by two areas, each area including a solid layer type structure substantially transparent to a light an optical birefringent device disposed between said pair of substrates

LEVEL 1 - 63 OF 225 PATENTS

5,169,754

Dec. 8, 1992

Biodegradable particle coatings having a protein covalently immobilized by means of a crosslinking agent and processes

for making same

INVENTOR: Siiman, Olavi, Davie, Florida Burshteyn, Alexander, Miami Lakes, Florida Gupta, Ravinder K., Pembroke Pines, Florida

A, acid cured gelatin of Bloom in the range 60 to 300, and said layers on the individual particles being crosslinked by the action of a chemical crosslinking agent such that aid particles can be ... with two layers of water soluble gelatin having a plurality of pendant functional groups, said gelatin layers comprising a first layer of type B, alkali cured gelatin of Bloom in the range 60 to 225 and a second layer of type ... [\*1] particles in which each particle comprises a solid core coated

[\*27] colloidal sized solid core material;

- to 225 and a second layer of type A, acid cured gelatin of Bloom in the range of 60 to 300; (b) a gelatin coating adsorbed onto the surface of said solid core and crosslinked thereon by a chemical crosslinking agent, said gelatin coating comprising first layer of type B, alkali cured gelatin of Bloom in the range 60
- (c) am antibody; and
- (d) a bridging group having an end covalently bonded to said crosslinked gelatin surface and another end covalently ...
- an antibody covalently bound to the surface of a crosslinked gelatin coated solid core particle wherein said gelatin coating comprises a first layer of type B, alkali cured gelatin of Bloom in the range 60-225 and a second layer of type A, acid cured gelatin of Bloom in the range of 60-300;
  - (b) incubating the mixture of step (a) for a time and at a temperature sufficient to insure the formation of a complex between  $\dots$

LEVEL 1 - 64 OF 225 PATENTS

5,162,782

## =2> GET 1st DRAWING SHEET OF

Nov. 10, 1992

Display device with coordinate input function

INVENTOR: Yoshioka, Kazuo, Nagasaki, Japan

for both displaying images and inputting coordinates, comprising: ... [\*1] a sensor means for sensing coordinate input detection signals from a control means: a liquid crystal display panel of two-layer type including an optical phase liquid crystal compensation cell as a first layer of the liquid crystal display panel and a liquid crystal display cell as a second layer of the liquid ...

... [\*1] signals to said sensor means.

[\*2] 2. A display device with coordinate input function as set forth in claim 1, wherein an image display screen of the liquid crystal display panel of two-layer type is divided into plural areas to be separately driven. PAGE 772

LEVEL 1 - 65 OF 225 PATENTS

5,148,259

<=2> GET 1st DRAWING SHEET OF 13

Sep. 15, 1992

Semiconductor device having thin film wiring layer of aluminum containing carbon

INVENTOR: Kato, Takashi, Sagamihara, Japan Ito, Takashi, Kawasaki, Japan

ito, lakasni, kawasaki, Ja Maeda, Mamoru, Tama, Japan carbon greater than an atomic percent of carbon contained in said ... [\*8] third layer. [\*9] 9. A semiconductor device as claimed in claim 5 in which a plurality of pairs of said third layer type and said fourth layer type are provided on said fourth layer in alternate succession so that each third layer type is sandwiched between two fourth layer types.

 $[*10] \,\, 10.$  A semiconductor device as claimed in claim 1, in which grains of said second layer are generally oriented on a (200) plane.

[\*11]

## GET 1st DRAWING SHEET OF 12

### Sep. 8, 1992

Neural net using capacitive structures connecting output lines and differentially driven input line pairs

INVENTOR: Engeler, William E., Scotia, New York

... [\*14] said training period of time for generating an error signal identified by the same ordinal number as said processor generating it.

[\*15] 15. A plurality, L in number, of neural net layers of the type set forth in claim 14, respectively identified by consecutive ordinal numbers zeroeth through (L - 1), L being a positive integer, the output ports of the processors in ... ... [\*26] input lines and the one of said (M+1) and 2M input lines identified by ordinal number M higher.

[\*27] 27. A plurality, L in number, of neural net layers of the type set forth in claim 25, respectively identified by consecutive ordinal numbers zeroeth through (L - 1), L being a positive integer, the non-linear amplifiers of the processors ... ... [\*31] lines that are identified by the same ordinal number connected to be receptive of the same input signal, thereby to provide a neural network

[\*32] 32. A plurality, L in number, of neural net layers of the type set forth in claim 31, respectively identified by consecutive ordinal numbers zeroeth through (L - 1), L being a positive integer, the non-linear amplifiers of the processors ...

 $\dots$  [\*33] said training period of time for generating an error signal identified by the same ordinal number as said processor generating it.

[\*34] 34. A plurality, L in number, of neural net layers of the type set forth in claim 33, respectively identified by consecutive ordinal numbers zeroeth through (L - 1), L being a positive integer, the output ports of the processors in ...

LEVEL 1 - 67 OF 225 PATENTS

5,141,684

0 GET 1st DRAWING SHEET OF

PAGE

Method of preparing urethane foam articles

INVENTOR: Yoshimura, Kimio, Urawa, Japan Narumi, Satoshi, Tokyo, Japan

What is claimed is:

1. A method of preparing a urethane foam article having a high density [\*1] 1. A method of preparing a urethane foam article having a high outer surface layer, of the type wherein a plastic liquid containing isocyanates, polyols, a catalyst, a blowing agent, an assistant and other additives is poured into a mold through a one-shot molding process while maintaining the ...

... [\*4] 1-substituted imidazole compound, 1,8-diazabicyclo(5,4,0)-7-undecene and an organic acid salt thereof.

[\*5] 5. A method of preparing a urethane foam article having a high density outer surface layer, of the type wherein a plastic liquid containing isocyanates, polyols, a catalyst, a blowing agent, an assistant and other additives is poured into a mold through a one-shot molding process while maintaining the ...

LEVEL 1 - 68 OF 225 PATENTS

5,132,132

<=2> GET 1st DRAWING SHEET OF

3

Jul. 21, 1992

Coating method for magnetic recording medium

INVENTOR: Watanabe, Masaru, Nishinomiya, Japan Hirose, Satoshi, Amagasaki, Japan

and then coating on said lower layer a second magnetic coating solution so as to form an upper layer thereon to thereby produce a two-layer type magnetic recording medium, said method comprising the steps of coating said first magnetic coating solution on said support by a first die including first and as to form a lower layer on said support continuously travelling second lip portions which are in ...

LEVEL 1 - 69 OF 225 PATENTS

5,128,229

<=2> GET 1st DRAWING SHEET OF

Jul. 7, 1992

Electrophotosensitive material and method of manufacturing

PAGE 75

GE 76

NVENTOR: Katsukawa, Masato, Ibaraki, Japan Tsujita, Mitsuji, Osaka, Japan Miura, Satoru, Shijonawate, Japan Kimoto, Keizo, Hirakata, Japan

We claim:

- the photosensitive layer includes a charge generating material a polycarbonate resin as a binding resin, said polycarbonate resin being ... photosensitive layer formed on the surface of a conductive substrate, wherein 1. An electrophotosensitive material having a single-layer type
- ... [\*2] according to claim 1, wherein the photosensitive layer includes a perylene compound as the charge generating material..
- [\*3] 3. A method of manufacturing an electrophotosensitive material, said electrophotosensitive material being a single-layer type photosensitive layer formed on the surface of a conductive substrate, wherein the photosensitive layer a charge generating material includes a polycarbonate resin as a binding resin, said polycarbonate resin being ...
- ... [ $^{\#5}$ ] compound represented by formula (II) is includes in an amount of 40 to 200 parts by weight per 100 parts by weight of the polycarbonate resin.
- [\*6] 6. An electrophotosensitive material having a single-layer type photosensitive layer formed on the surface of a conductive substrate, wherein the photosensitive layer includes a charge generating material a polycarbonate resin represented by the following formula (I): [See Original Patent ...
- said photosensitive layer being not greater than 2.5 x 10< 3 ... [\*6] mu 1/mg.
- [\*7] 7. A method of manufacturing an electrophotosensitive material, said electrophotosensitive material being a single-layer type photosensitive layer formed on the surface of a conductive substrate, wherein the photosensitive layer a charge generating material includes a polycarbonate resin represented by the following formula (I): [See Original Patent ...

5,126,210

0 GET 1st DRAWING SHEET OF <=5>

Jun. 30, 1992

Anodic phosphonic/phosphinic acid duplex coating on valve metal surface

INVENTOR: Wieserman, Larry F., Apollo, Pennsylvania Wefers, Karl, Apollo, Pennsylvania

Gary A., Natrona, Pennsylvania dward S., New Kensington, Pennsylvania

copper, manganese, molybdenum, chromium, nickel, zinc, vanadium, boron, lithium and zirconium; and

duplex layer comprised of:

intermediate layer consisting essentially of a non-porous barrier e valve metal oxide attached to said base layer; and

n acid resistant, functionalized layer of a monomeric s-containing compound chemically bonded to a surface of said oxide e functionalized ...

9. A layered material comprised of:

base layer of aluminum alloy; and

duplex layer comprised of:

intermediate layer consisting essentially of a non-porous barrier e aluminum oxide attached to said base layer; and

n acid resistant, functionalized layer of an organic monomeric s-containing compound chemically bonded to a surface of said oxide

10. The layered ...

31] A layered material comprised of:

base layer of aluminum or aluminum alloy; and

duplex layer comprised of:

intermediate layer consisting essentially of a non-porous barrier e aluminum oxide attached to said base layer having a density of 2.8 to c. being greater than 95 wt. % aluminum oxide, having a thickness in of 100 to 5000Angstrom and ... LEVEL 1 - 71 OF 225 PATENTS

5,116,692

N GET 1st DRAWING SHEET OF

May 26, 1992

Multi-layer type sliding bearing of aluminum alloy and method of producing the same

(i) an layer typ (ii) a phosphoru layer, th ... [\* (a) a (b) a (i) an layer typ (ii) a phosphoru layer. [\*10] ... [\* (a) a (b) a (b) a (b) a (b) a (i) an a (b) a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i) an a (i)

What is claimed is:

- [\*1] 1. In a multi-layer type sliding bearing of aluminum alloy, having a backing layer of steel, a bearing layer of aluminum alloy bonded to the backing layer, and an overlay layer, the improvement further comprising a mixture layer of a ...
- ... [\*1] layer and one element selected from the group consisting of Ni, Co and Fe, said overlay consisting by weight of 0 to 15% Cu, 0 to 20% Sb, and the balance- Sn and incidental impurities.
- [\*2] 2. A multi-layer type sliding bearing of aluminum alloy, having a backing "layer of steel provided on one side thereof with a rear face-plating layer provided on rear face thereof, a bearing layer of aluminum alloy bonded to
- ... [42] overlay and one element selected from the group consisting of Ni, Co and Fe, said overlay consisting by weight of 0 to 15% Cu, 0 to 20% Sb, and the balance Sn and incidental impurities.
- [\*3] 3. A multi-layer type sliding bearing of aluminum alloy as claimed in claim 2, wherein the rear face-plating layer consists of the same constituents as the overlay, the thickness of the rear face-plating layer being in a range of 0.1 to 5 microns.
- [\*4] 4. In a method of producing a multi-layer type sliding bearing of aluminum alloy, comprising the steps of: providing a half cylindrical or cylindrical bearing member made of an aluminum alloy, said bearing member being bonded onto a backing layer of steel; and ...
- ... [\*4] said mixture layer being constituted by a mixture of the constituents of said overlay and one element selected from the group consisting of Ni, Co and Fe.
- [\*5] 5. A method of producing a multi-layer type sliding bearing of aluminum alloy claimed in claim 4, wherein said step of electrolytically providing said overlay layer on said inner face of said bearing member further includes the step of electrolytically ...

LEVEL 1 - 72 OF 225 PATENTS

5,103,329

<=2> GET 1st DRAWING SHEET OF 8

Apr. 7, 1992

# Surface stabilized ferroelectric liquid crystal switching using proximity effects

INVENTOŘ: Clark, Noel A., Boulder, Colorado Handschy, Mark, Boulder, Colorado

third electrode means, said first and second domain wall forming a data Vallwe. [\*15] 15. The apparatus as recited in claim 1, wherein said liquid crystal film is a tilted layer type.

[\*16] 16. An apparatus as in claim 1, wherein said electrode gap is spanned by a resistive layer.

80

PAGE

LEVEL 1 - 73 OF 225 PATENTS

5,087,544

Feb. 11, 1992

Perylene electrophotosensitive material with m-phenylenediamine

NVENTOR: Muto, Nariaki, Daito, Japan Sumida, Keisuke, Hirakata, Japan Nakazawa, Toru, Osaka, Japan Matsumoto, Kazuo, Hirakata, Japan Kakui, Mikio, Mino, Japan

What is claimed is:

[\*1] 1. An electrophotosensitive material comprising a conductive substrate and a single layer type photosensitive layer provided on said conductive substrate, said photosensitive layer containing a m-phenylenediamine compound as charge-transferring material and a perylene compound as charge-generating

LEVEL 1 - 74 OF 225 PATENTS

5,081,513

S GET 1st DRAWING SHEET OF

Jan. 14, 1992

Electronic device with recovery layer proximate to active layer

INVENTOR: Jackson, Warren B., San Francisco, California Hack, Michael, Mountain View, California

...  $[\# \S]$  dopant concentration of each of said dopant types in said recovery layer is capable of providing substantially the same number of carriers of said

opposite sign to said channel carriers as does a doped layer of that type having a concentration in the range of 1.5 x 10<18 > to 4.5 x 10<18 > atoms per cm<3> .

[\*10] 10. An electronic device including a substantially intrinsic non-single ...

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PAGE

LEVEL 1 - 75 OF 225 PATENTS

5,059,502

GET 1st DRAWING SHEET OF

Oct. 22, 1991

Electrophotographic photoconductor

INVENTOR: Kojima, Narihito, Numazu, Japan Yamazaki, Shunpei, Atsugi, Japan Hayashi, Shigenori, Atsugi, Japan **Fakeyama**, Junichi, Atsugi, Japan Nagame, Hiroshi, Numazu, Japan Seto, Mitsuru, Yamakita, Japan Ishida, Noriya, Atsugi, Japan Hirose, Naoki, Atsugi, Japan Sasaki, Mari, Atsugi, Japan

... [\*16] 1, wherein the Vickers hardness of said organic photoconductive layer is 10 to 50 Kg/mm<2> .

[\*17] 17. The electrophotographic photoconductor as claimed in claim 1, wherein organic photoconductive layer is a single-layer-type photoconductive

[\*18] 18. The electrophotographic photoconductor as claimed in claim 17, wherein the thickness of said single-layer-type photoconductive layer is 5 to 30  $\,$ 

[\*19]. 19. The electrophotographic photoconductor as claimed in claim 1, wherein bryanic photoconductive layer is a function-separated-type photoconductive layer comprising a ... LEVEL 1 - 76 OF 225 PATENTS

5,054,134

2 GET 1st DRAWING SHEET OF

Oct. 8, 1991

Upper layer water flow type circulating water pool

INVENTOR: Teratsuji, Osamu, Ichikawa, Japan Nishimura, Keiichi, Urawa, Japan

PAGE

83

Moriya, Yoshiro, Matsudo, Japan Ueda, Yukihiko, Yokohama, Japan

What is claimed is:

[\*1] 1. An upper water flow layer type circulating water pool comprising a circulating pool main body with front and rear curved portions; a swimming tank or pool defined by an opening in an upper portion of said circulating ...

PAGE

LEVEL 1 - 77 OF 225 PATENTS

5,051,126

<=2> GET 1st DRAWING SHEET OF 1

Sep. 24, 1991

Cermet for tool

INVENTOR: Yasui, Hajime, Nagoya, Japan Suzuki, Junichiro, Hashima, Japan group consisting of the group IVb metals than the core, and the core is composed of more transitional metals selected from the group consisting of the group Vb metals and tungsten than any outer layer of the Type-II particles.

[\*2] 2. The cermet of claim 1, wherein the ratio of transitional metals in group IVb, transitional metals in group Vb, and tungsten to carbon and nitrogen is 1.0:0.85-1.0.

[\*3] 3. The cermet of claim ... LEVEL 1 - 78 OF 225 PATENTS

5,050,323

<=2> GET 1st DRAWING SHEET OF 2

Sep. 24, 1991

Badge

INVENTOR: Gagnon, Raymond, Montreal, Canada

... [\*1] front face of said body layer, and

fastening means carried by the back face of said body layer to attach said body layer to a wearer's clothing; wherein:

the foam material forming the body layer is of the type having no memory when

said foam material is left exposed at the peripheral edge of said body layer;

said badge further comprises a protecting transparent film covering said image-béaring ...

... [\*9] front face of said body layer; and,

fastening means carried by the back face of said body layer to attach said body layer to a wearer's clothing; wherein:

the expanded polystyrene forming the body layer is of the type having no memory when compressed; said expanded polystyrene is left exposed at the peripheral edge of said body

said badge further comprises a protecting transparent film covering said image-bearing ...

... [\*10] front face of said body layer; and,

fastening means carried by the back face of said body layer to attach said body layer to a wearer's clothing, wherein;

the expanded polystyrene forming the body layer is of the type having no memory when compressed;

said expanded polystyrene is left exposed at the peripheral edge of said body layer; and,

said badge further comprises a protecting transparent film covering said image-bearing ...

LEVEL 1 - 79 OF 225 PATENTS

5,039,627

GET 1st DRAWING SHEET OF

Aug. 13, 1991

Method of producing a quasi-flat semiconductor device capable of a multi-wavelength laser effect and the corresponding device

INVENTOR: Menigaux, Louis, Bures sur Yvette, France Dugrand, Louis, Chelles, France

gallium and indium and at least one material selected from the group consisting of aluminum, of phosphorous, arsenic and antimony.

- [\*5] 5. A method according to claim 4, wherein the active layers are of type N, and comprise Ga 1-x Al x As, x differing each time, and being less than approximately 10%, while the confining layers comprise Ga 1-y Al y ...
- ... [\*9] about one-tenth of a micron.
- 10. A method according to claim 1, wherein the upper layer is a group [\*10] 16 III-V alloy.
  - $[\star 11]$  11. A method according to claim 5, wherein the upper layer is type gallium arsenide.
- [\*12] 12. A method according to claim 1, wherein said levelling comprises an erosion operation applied to the upper surface of the block, laying bare at least over the major part of the eroded ...
- ... [\*30] least one material selected from the group consisting of aluminum, gallium and indium and at least one material selected from the group consisting of phosphorous, arsenic and antimony.
- [\*31] 31. A method according to claim 30, wherein the active layers are of type N, and comprises Ga 1-x Al x As, x differing each time and being less than approximately 10%, while the confinement layers comprise Ga 1-y Al y ...

LEVEL 1 - 80 OF 225 PATENTS

5,037,505

<=2> GET 1st DRAWING SHEET OF 2

Aug. 6, 1991

Construction process for a self-aligned transistor

INVENTOR: Tung, Pham N., Paris, France

- ... [\*2] mask,
- i) dissolving the two silica masks in a solution of HF + NH4F + H2O.
- [\*3] 3. Process of constructing a self-aligned transistor, according to claim 1, from a substrate comprising layers of type N and N< + > formed by epitaxy, further comprising the steps of:
- a) depositing and masking by a first resin mask creating a resin pattern of dimensions corresponding to the power of resolution of said ...

LEVEL 1 - 81 OF 225 PATENTS

5,031,025

=2> GET 1st DRAWING SHEET OF 3

## Jul. 9, 1991

Hermetic single chip integrated circuit package

INVENTOR: Braun, Robert E., Norristown, Pennsylvania Gibbs, Ronald T., King of Prussia, Pennsylvania

- ... [\*1] said rim of said lid being sealed to said substrate, thereby forming an hermetic package.
- [\*2] 2. An hermetic integrated circuit package as defined in claim 1 wherein said wiring substrate is of the multi-layer type, said lead terminals being coupled to said input/output terminals.
- [\*3] 3. An hermetic integrated circuit package as defined in claim wherein said rim of said lid is comprised of a ... LEVEL 1 82 OF 225 PATENTS

5,028,786

GET 1st DRAWING SHEET OF <=5>

Jul. 2, 1991

Array for a nuclear radiation and particle detector

INVENTOR: Da Silva, Angela J., Vancouver, Canada Le Gros, Mark A., Vancouver, Canada Turrell, Brian G., Vancouver, Canada Kotlicki, Andrzej, Warsaw, Maryland, Poland Drukier, Andrzej K., Greenbelt, Maryland

detector as defined in claim 8 wherein said each array is planar ārray. ... [\*13]

[\*14] 14. A method of making a detector array comprising depositing a substantially continuous film layer of type I superconducting material on a substrate removing a portion of said film to leave a plurality of discrete pixels each of a predetermined size of said type I superconducting material

LEVEL 1 - 83 OF 225 PATENTS

5,028,505

Jul. 2, 1991

Electrophotographic photoreceptor

INVENTOR: Akasaki, Yutaka, Kanagawa, Japan Nukada, Katsumi, Kanagawa, Japan Sato, Katsuhiro, Kanagawa, Japan

... [\*9] comprising a compound of formula (I), (II), or (III) as set forth in claim 1.

[\*10] 10. The electrophotographic photoreceptor as claimed in claim 1, wherein the photosensitive layer is of a single layer type. 91

LEVEL 1 - 84 OF 225 PATENTS

5,024,913

Jun. 18, 1991

Electrophotographic photosensitive material

INVENTOR: Yoshida, Takeshi, Kawachinagano, Japan Nakatani, Kaname, Osaka, Japan Fukami, Toshiyuki, Sakai, Japan Tanaka, Nariaki, Kishiwada, Japan

... [\*6] photosensitive layer.

[\*7] 7. The electrophotographic photosensitive material of claim 6, wherein the surface protective layer is 2 to 5 microns thick.

[\*8] 8. The electrophotographic photosensitive material of claim 1, wherein the photosensitive layer is of a single layer type.

[\*9] 9. The electrophotographic photosensitive material of claim 8, wherein the photosensitive layer is 10 to 50 microns thick.

[\*10] 10. The electrophotographic photosensitive material of claim 8, wherein the photosensitive layer is 15 to 25 microns thick.

[\*11] 11. The ...

LEVEL 1 - 85 OF 225 PATENTS

5,022,441

<=2> GET 1st DRAWING SHEET OF 67

Jun. 11, 1991

Papermaker's double layer fabric with high warp and weft volume per repeat

INVENTOR: Tate, Takuo, Hachiouji, Japan Watanabe, Taketoshi, Inagi, Japan Nagura, Hiroyuki, Inagi, Japan

What is claimed is:

- [\*1] 1. A papermakers' double layer type fabric comprising in one repeat warp layer, said warp layer having an upper surface and a lower surface, said warp layer consisting of n x 2 of warps wherein n is an ...
- ... [\*1] being each interlaced once in one repeat with a warp, wherein said lower surface polyamide wefts are of larger diameter than said lower surface polyester wefts.
- [\*2] 2. A papermakers' double layer type fabric comprising in one repeat warp layer, said warp layer having an upper surface and a lower surface, said warp layer consisting of n x 2 of warps wherein n is an  $\dots$
- ... [\*2] between which said non-interlacing warp is located to form a knuckle so that the knuckles so formed on each of the adjacent lower surface wefts are arranged in a staggered relation.
- [\*3] 3. A papermakers' double layer type fabric according to claim 2, wherein a non-interlacing warp sandwiched in between a pair of warps interlacing with a lower surface polymeric weft, interlaces with an upper surface weft at a position where said pair of warps interlace with the lower surface polymeric
- [\*4] 4. A papermakers' double layer type fabric according to claim 2, wherein in said n x 2 of warps, a non-interlacing warp and a warp interlacing with a lower surface polymeric weft, are alternately arranged.
- wherein in said n x 2 of warps, a non-interlacing warp and a plurality of warps interlacing with a lower surface polymeric weft, are alternately arranged. 5. A papermakers' double layer type fabric according to claim 2, [\*2]
- [\*6] 6. A papermakers' double layer type fabric according to claim 2, wherein said lower surface polyamide wefts are of larger diameter than said lower surface polyester wefts.
- ø [\*7] 7. A papermakers' double layer type fabric comprising in one repeat warp layer, said warp layer having an upper surface and a lower surface, said warp layer consisting of n x 2 of warps, wherein n is an ...
- ... [\*7] each interlaced once in one repeat with a warp and the lower surface polyester wefts being each interlaced twice in one repeat with a warp. PAGE 93

Pat. No. 5022441, \*7

- [\*8] 8. A papermakers' double layer type fabric according to claim 7, wherein said lower surface polyamide wefts are of larger diameter than said lower surface polyester wefts.
- [\*9] 9. A papermakers' double layer type fabric according to claim 7, wherein the number of said lower surface polyamide wefts and that of said lower surface polyester wefts are in a ratio of from 1:3 to 3:1.

- $[*10]\ 10.$  A papermakers' double layer type fabric according to claim 7, wherein both said lower surface polyamide wefts and lower surface polyester wefts are each interlaced with two adjacent warps.
- [\*11] 11. A papermakers' double layer type fabric according to claim  $1\theta$ , wherein said lower surface polyamide wefts are of larger diameter than said lower surface polyester wefts.
- [\*12] 12. A papermakers' double layer type fabric according to claim 10, wherein the number of said lower surface polyamide wefts and that of said lower surface polyester wefts are in a ratio of from 1:3 to 3:1.
- repeat with a warp, and said lower surface polyamide wefts are each interlaced  $[\star 13]$  13. A papermakers' double layer type fabric according to claim 7, wherein said lower surface polyester wefts are each interlaced twice in one
- ... [\*13] pair of adjacent warps between which a warp interlacing with an upper surface weft at a position where said pair of warps interlace with the lower surface polyamide weft, is disposed.
- [\*14] 14. A papermakers' double layer type fabric according to claim 13, wherein a non-interlacing warp is arranged adjacent to a warp interlacing with said lower surface polyester weft.
- [\*15] 15. A papermakers' double layer type fabric according to claim 13, wherein at least one of a pair of warps which interlace with said lower surface polyamide weft also interlaces with the lower surface polyester weft.
- [\*16] 16. A papermakers' double layer type fabric according to claim 13, wherein said lower surface polyamide wefts are of larger diameter than said lower surface polyester wefts.
- [\*17] 17. A papermakers' double layer type fabric according to claim 13, wherein the number of said lower surface polyamide wefts and that of said lower surface polyester wefts are in a ratio of 1:3 to 3:1.
- 18. A papermakers' double layer type fabric according to claim 13, wherein said lower surface polyamide wefts are each interlaced once in one repeat with a pair of adjacent warps between which a warp interlacing with an upper surface ... [\*18]
- warp located between a pair of warps interlacing with a lower surface polyamide polyester wefts are each interlaced twice in one repeat with a

Pat. No. 5022441, \*18

[\*19] 19. A papermakers' double layer type fabric according to claim 18, wherein said lower surface polyamide wefts are of larger diameter than said lower surface polyester wefts.

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wherein the number of said lower surface polyamide wefts and that of said lower surface polyester wefts are in a ratio of from 1:3 to 3:1.

LEVEL 1 - 86 OF 225 PATENTS

5,005,057

GET 1st DRAWING SHEET OF 19

Apr. 2, 1991

Semiconductor light-emitting diode and method of manufacturing the same

Tokyo, Japan INVENTOR: Izumiya, Toshihide, Ohba, Yasuo, Yokohama, Japan Hatano, Ako, Tokyo, Japan ... [\*7] conductivity type having a zinc blend type crystal structure.

[\*8] 8. A diode according to claim 3 or 7, wherein said light-reflection layer has a multilayered structure in which said superlattice layers of the types are alternately stacked with a period which is substantially equal to the light-emitting wavelength.

[\*9] 9. A semiconductor light-emitting diode including a light-emitting layer having a p-n junction, comprising:

PAGE

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4,996,108

LEVEL 1 - 87 OF 225 PATENTS

6 GET 1st DRAWING SHEET OF

Feb. 26, 1991

Sheets of transition metal dichalcogenides

INVENTOR: Divigalpitiya, W. M. Ranjith, Vancouver, Canada Frindt, Robert F., Vancouver, Canada Morrison, S. Roy, Burnaby, Canada

What is claimed is:

[\*1] 1. A process for forming sheet-like compositions of the formula:

MX2:Y

wherein MX2 is a layer-type transition metal dichalcogenide, M is a metal selected from the group consisting of niobium, tantalum, molybdenum and

... [\*18] A method of coating an object, comprising:

forming a sheet-like composition at an interface between water and a non-metallic liquid which is immiscible with water, the composition having the

X2.V

wherein MX2 is a layer-type transition metal dichalcogenide, the M is selected from the group consisting of niobium, tantalum, molybdenum and tungsten; X is selected from the group consisting of sulfur and selenium, and Y is a material located between layers of MX2; and

bringing the ...

... [\*18] spread over the object.

[\*19] 19. A method as claimed in claim 18, wherein the MXZ is crystalline with c-axes perpendicular to the substrate.

[\*20] 20. A composition having the formula:

MX2:Y

wherein MX2 is a layer-type transition metal dichalcogenide selected from the group consisting of molybdenum disulfide and tungsten disulfide; and Y is an PAGE 97

Pat. No. 4996108, \*20

organic material located between layers of MX2.

[\*21] 21. A composition as claimed in claim 20, wherein the ...

... [\*24] tetrachloride; dimethoxy benzene; 1-chloronaphthalene; chrysene; stearamide; phthalocyanine; copper phthalocyanine and iron pentacarbonyl.

25. A object having a surface coated with a composition having the [\*25]formula:

MX2:Y

wherein MX2 is a layer-type transition metal dichalcogenide, M is a metal selected from the group consisting of niobium, tantalum, molybdenum and tungsten, X is a chalcogen selected from the group consisting of sulfur and selenium, and Y is an organic substance located ... LEVEL 1 - 88 OF 225 PATENTS

717

## <=2> GET 1st DRAWING SHEET OF 2

Dec. 25, 1990

Method of producing a semiconductor laser

INVENTOR: Takahashi, Shogo, Itami, Japan

What is claimed is:

 $[*1]_{\mathbb{R}^2}$  1. A method of producing a semiconductor laser comprising:

growing at least a p type lower cladding layer, a quantum well active layer, and an type upper cladding layer successively on a substrate;

depositing a first film as a source for diffusion of n type impurities on portion of the n type upper cladding layer;

LEVEL 1 - 89 OF 225 PATENTS

99

PAGE

4,980,216

<=2> GET 1st DRAWING SHEET OF 2

Dec. 25, 1990

Transfer for textiles

INVENTOR: Rompp, Walter, Rosenstrasse 46, D-7406 Mossingen, Federal Republic of Germany

... [\*10] freely cross-linkable silicon emulsion.

[\*11] 11. A transfer according to claim 10, wherein said separating layer is a polysiloxane compound.

[\*12] 12. A transfer according to claim 1, wherein said separating layer is of the type C1G 2 pure.

 $[^{*}14] \ \ 14.$  A transfer according to claim 1, wherein said separating layer is manufactured on a ... [\*13] 13. A transfer according to claim 1, wherein said separating layer is screen-printable.

LEVEL 1 - 90 OF 225 PATENTS

4,976,990

Dec. 11, 1990

Process for metallizing non-conductive substrates

INVENTOR: Bach, Wolf, Southbury, Connecticut Ferrier, Donald R., Thomaston, Connecticut Kukanskis, Peter E., Woodbury, Connecticut Williams, Ann S., Southbury, Connecticut Senechal, Mary J., Canton, Connecticut

<u>\_</u>

- adherently and essentially void-free onto said catalyzed through-hole surfaces. electroless depositing solution to deposit metal fully and
- [\*4] 4. In a process for manufacturing printed circuit boards of the multilayer type, in which a planar composite substrate material is provided comprised of a laminate of alternating parallel layers of metal and non-conductive, glass-reinforced thermosetting or thermoplastic material, and in which through-holes are provided ...
- metal depositing solution to deposit metal fully and adherently and essentially void-free onto said catalyzed through-hole surfaces. [\*4]
- layer type, in which a planar composite substrate material is provided comprised of a laminate of alternating parallel layers of metal and non-conductive, glass-reinforced thermosetting or thermoplastic material, and in which 5. In a process for manufacturing printed circuit boards of the multithrough-holes are provided ...
- ... [\*11] time of said process for metallizing said through-hole surfaces.
- [\*12] 12. A process for providing metallized through-holes in a printed circuit board of the double-sided or multi-layer type, comprising the steps of
- (a) providing a printed circuit substrate material comprised of a member selected from the group consisting of (1) a planar non-conductive material comprised of glass-reinforced thermosetting or ...
- ... [\*12] metal depositing solution to deposit metal fully and adherently and essentially void-free onto said catalyzed through-hole surfaces.
- [\*13] 13. The process according to claim 12 wherein said printed circuit board is of the multi-layer type and wherein said through-hole surfaces are desmeared between steps (b) and (c).
- [\*14] 14. A process for providing a full-coverage, essentially void-free, adherent metal layer on the surface of a ... LEVEL 1 91 OF 225 PATENTS

4,963,450

Oct. 16, 1990

Electrophotographic photosensitive member with disazo pigment

INVENTOR: Miyazaki, Hajime, Yokohama, Japan Go, Shintetsu, Yokohama, Japan Senoo, Akihiro, Yokohama, Japan Iuchi, Kazushi, Yokohama, Japan Kanemaru, Tetsuro, Tokyo, Japan

What is claimed is:

[\*1] 1. A laminated layer type electrophotographic photosensitive member having a charge generation layer and a charge transport layer on an electroconductive support, characterized in that the charge generation layer has at least one of disazo pigments [is by] of the formulae (1) and (... PAGE 102

LEVEL 1 - 92 OF 225 PATENTS

4,941,737

<=2> GET 1st DRAWING SHEET OF 5

Jul. 17, 1990

Liquid-crystal display device using twisted nematic liquid crystal molecules

INVENTOR: Kimura, Naofumi, Nara, Japan

What is claimed is:

[\*1] 1. A liquid-crystal display device comprising:

a multi-layer-type liquid-crystal cell that is composed of at least first and second cell layers, said cell layers containing liquid-crystal molecules with a twisted nematic orientation therein;

said first cell layer having an angle of ... LEVEL 1 - 93 OF 225 PATENTS

4,932,788

<=2> GET 1st DRAWING SHEET OF 4

Jun. 12, 1990

Monitoring of the quality of a flowing vapor

INVENTOR: Yeh, George C., 2 Smedley Dr., Newtown Square, Pennsylvania 19073

... [\*5] 1 wherein said flowmeter is a mass flowmeter capable of directly metering the mass flow rate of the vaporized sample.

 $[\star 6]$  6. A system according to claim 5 wherein said mass flowmeter is of an electrothermal boundary-layer type flowmeter in which temperature sensing and

heating elements are placed outside the pipe carrying said stream of sample vapor and do not obstruct the stream.

- [\*7] 7. A system according to claim 1 wherein said means ...
- ... [\*16] said heater means further comprises:

a thermal insulator completely covering said shell and the inlet and outlet

[\*17] 17. Apparatus according to claim 11 wherein said mass flowmeter means is of the electrothermal boundary-layer type for permitting unobstructed flow.

[\*18] 18. Apparatus according to claim 11 wherein said second sensing means comprises.

thermistors formed into thin bands and placed around the inlet and outlet of said heater means.

LEVEL 1 - 94 OF 225 PATENTS

[\*19]

4,888,261

<=2> GET 1st DRAWING SHEET OF 5

Dec. 19, 1989

Electrophotographic photosensitive member

INVENTOR: Mabuchi, Minoru, Tokyo, Japan

... [\*6] electrophotographic photosensitive member of claim 1, wherein said charge transport layer is laminated on said charge generation layer.

[\*7] 7. The electrophotographic photosensitive member of claim 1, wherein said photosensitive layer has a single layer type that the charge-generating material and the charge-transporting material are contained in the same layer. PAGE 105

LEVEL 1 - 95 OF 225 PATENTS

4,886,721

<=2> GET 1st DRAWING SHEET OF 10

Dec. 12, 1989

Electrophotographic plate by use of metal naphthalocyanine derivative

INVENTOR: Hayashida, Shigeru, Hitachi, Japan Tai, Seiji, Hitachi, Japan

uerryaciya. OR: Hayashida, Shigeru, Hitachi, Japan

(GE 104

Hayashi, Nobuyuki, Hitachi, Japan Iwakabe, Ya≲ushi, Hitachi, Japan Kinjo, Noriyuki, Hitachi, Japan Numata, Shunichi, Hitachi, Japan ... [\*3] germaniumnaphthalocyanine,

bis(tripropylsiloxy)germaniumnaphthalocyanine,

bis(tributylsiloxy)germaniumnaphthalocyanine,

bis(triphenylsiloxy)germaniumnaphthalocyanine and

bis(triethylsiloxy)tinnaphthalocyanine.

[\*4] 4. The electrophotographic plate according to claim 1, wherein said photoconductive layer is a complex double layer type comprising a charge generation layer containing said metal naphthalocyanine derivative which is a charge generation substance, and said charge transport layer containing a charge transport substance.

\*5] 5. The electrophotographic ... LEVEL 1 - 96 OF 225 PATENTS

4,877,702

Oct. 31, 1989

Electrophotographic sensitive material

INVENTOR: Miyamoto, Eiichi, Osaka, Japan Mutou, Nariaki, Daito, Japan

Makazawa, Tooru, Osaka, Japan Nakazawa, Tooru, Osaka, Japan

What is claimed is:

characterized in that said electric charge generating substance is a perylene [\*1] 1. An electrophotographic sensitive material provided with a single-layer type sensitive layer containing an electric charge generating substance, an electric charge transferring substance, and a binding resin, type compound ...

LEVEL 1 - 97 OF 225 PATENTS

107

1

4,867,827

Sep. 19, 1989

Process for gold foil stamping in relief

INVENTOR: Lesieur, Frederic, 25, Rue Pradier, Paris, France 75019

I claim:

[\*1] 1. A method of gilding raised images formed by a thermographic process on a substrate with a marking layer of the type releasably disposed on a backing film comprising the steps of:

providing a thermally activated adhesive powder of the type that exhibits adhesive properties while in a solidified state;

printing a selected ...

... [\*9] percentage basis, of 65% styrene oleophthalic resin; 15% to 20% acrylic resin; 10% to 15% plasticizer; 5% to 10% microcrystalline wax.

[\*10] 10. A method of gilding a substrate with a marking layer of the type releasably disposed on a backing film, comprising the steps of:

providing a thermally activated adhesive powder of the type that will liquefy under a sufficient amount of heat and will exhibit adhesive properties when ...

LEVEL 1 - 98 OF 225 PATENTS

4,852,693

2 <=2> GET 1st DRAWING SHEET OF

Aug. 1, 1989

Lubrication monitoring apparatus for machine

INVENTOR: Nakajima, Yoshiaki, Yono, Japan Suzuki, Tadashi, Machida, Japan Wada, Yoneji, Urawa, Japan

a separate pump and supplied to said lubricated mechanical ... [\*4] section. [\*5] 5. A lubrication monitoring apparatus for a machine as claimed in claim 1, wherein said filter is a deep-layer type filter.

 $[\star 6]$  6. A lubrication monitoring apparatus for a machine as claimed in claim 1, wherein said warning device is connected with a recorder to make a recording when difference in

LEVEL 1 - 99 OF 225 PATENTS

4,822,590

GET 1st DRAWING SHEET OF

Apr. 18, 1989

Forms of transition metal dichalcogenides

INVENTOR: Morrison, S. Roy, Burnaby, Canada Frindt, GRobert F., Vancouver, Canada Joensen, Per, Coquitlam, Canada Gee, Michael A., Vancouver, Canada [Miremadi, Bijan K., Coquitlam, Canada]

We claim:

1. An exfoliation process for preparing a single layer substance of  $\left[ egin{array}{c} *1 
ight] \ ext{the form} \end{array}$ 

**MX** 2

wherein MXZ is a layer type transition metal dichalcogenide selected from the group consisting of MoS2, TaS2, WS2, which comprises:

(a) intercalating multi-layer MX2 with an alkali metal in a dry environment for sufficient time to enable the ...
LEVEL 1 - 100 OF 225 PATENTS

110

PAGE

4,800,568

GET 1st DRAWING SHEET OF 1

Jan. 24, 1989

Gas laser with a frequency-selective dielectric layer system

INVENTOR: Krueger, Hans, Munich, Federal Republic of Germany Good, Hans P., Sargans, Switzerland

said means being a frequency selective layer system being formed ... [\*1] said means being a frequency selecon the Brewster window, said system comprising: a plurality of successive layers, said plurality of layers including only two layer types and including alternating high refractive index first layers and low refractive index second layers, each of said first layers having substantially a first optical thickness and each of said second layers having ...

 $\ldots$  [\*9] opposite ends through which laser radiation is emitted, an improved frequency selective layer system comprising:

layers including only two layer types and including alternating high refractive index first layers and low refractive index second layers, each of said first layers having substantially a thickness of 71.2 nm and each of said second a plurality of successive layers on said Brewster window, said plurality of ayers having ... 111

LEVEL 1 - 101 OF 225 PATENTS

Dec. 13, 1988

Mixed metal hydroxide-clay adducts as thickeners for water and other hydrophylic fluids

INVENTOR: Burba, III, John L., Angleton, Texas
Barnes, Audrey L., Lake Jackson, Texas

- ... [\*16] saconite, vermiculite, chlorite, attapulgite, sepiolite, palygorskite, and Fullers's earth.
- [\*17] 17. The method of claim 1 wherein the mineral clay is at least one of the group consisting of amorphous clays of the allophane group and crystalline clays of the 2-layer type, 3-layer type, expanding type, non-expanding type, elongate type, regular mixed layer type, and chain structure type.
- 18. The method of claim 1 wherein the mineral clay is bentonite. [\*18]
- 19. The method of claim 1 wherein the mineral clay is beneficiated bentonite [\*19]
- 20. The method of claim 1 wherein the weight ratio of monolayered [\*20]

112

4,775,814

LEVEL 1 - 102 OF 225 PATENTS

m GET 1st DRAWING SHEET OF

Oct. 4, 1988

Saw device

INVENTOR: Yuhara, Akitsuna, Kawasaki, Japan Hirashima, Tetsuya, Yokohama, Japan Sasaki, Jun, Sagamihara, Japan famada, Jun, Yokohama, Japan

- substrate, at least one set of finger electrodes disposed on said substrate, at least one set of said electrodes having a laminated structure made up of a plurality of layers including at least one of a first layer type of an aluminum film and at least one of a second layer type of an includes an impurity selected from the group consisting of titanium, chromium, vanadium and manganese.
- [\*17] 17. A SAW device according to claim 16, wherein said laminated structure includes a first layer of said first layer type formed on said substrate and a second layer of said second layer type formed on said first layer, the thickness of said first layer being greater than the thickness of said second layer.

substrate and a second layer of said first layer type formed on said first layer, the thickness of said second layer being greater than the thickness of [\*18] 18. A SAW device according to claim 16, wherein said laminated structure includes a first layer of said second layer type formed on said

[\*19] 19. A SAW device according to claim 16, wherein said laminated structure includes a first layer of said first layer type formed on said substrate, a second layer of said second layer type formed on said first layer, and a third layer of said first layer type formed on said second layer, the combined thickness of said first and third layers being greater than the thickness of said second layer. [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] [\*20] first and third layers. [\*21] 21. A SAW device according to claim 16, wherein said laminated structure includes a first layer of said first layer type formed on said substrate, a second layer of said second layer type formed on said first layer, a third layer of said first layer type formed on said second layer, and a fourth layer of said second layer type formed on said third layer, the combined thickness of said first and third layers being greater than the combined said second and fourth layers. Pat. No. 4775814, \*21 thickness of

 $[*22] \\ 22.$  A SAW device according to claim 17, wherein said first layer type includes an impurity selected from the group consisting of titanium, copper, magnesium, zinc and nickel.

23. A SAW device according to claim 22, wherein said first layer includes first and second sublayers having different ... LEVEL 1 - 103 OF 225 PATENTS

4,773,074

GET 1st DRAWING SHEET OF

Sep. 20, 1988

Dual mode laser/detector diode for optical fiber transmission lines

INVENTOR: Hunsperger, Robert G., Newark, Delaware Park, Jung H., Newark, Delaware

113

... [412] conductivity type of the active layer, the waveguide layer having first and second surfaces, the first surface in contact with the second surface of the active layer; a confining layer having a conductivity layer opposite the active layer type and having a surface in contact with the second surface of the waveguide layer;

cap and substrate layers; and

means to couple the cell and optical and electrical circuits. AGE 115

LEVEL 1 - 104 OF 225 PATENTS

4,761,242

<=2> GET 1st DRAWING SHEET OF 2

Aug. 2, 1988

Piezoelectric ceramic composition

INVENTOR: Suzuki, Kazunori, Nagoya, Japan Naitoh, Masataka, Kariya, Japan ... [\*14] constant-temperature characteristic, said additive being selected from the group consisting of 10-55 mol \$ CaTi03 and 1-15 mol \$ SrTi03, and

the balance of the composition being PbBi4Ti4015 having a Bi-layer type structure which PbBi4Ti4015 has a positive dielectric constant-temperature characteristic, a change in the dielectric constant of the PbBi4Ti4015 which change occurs by a change in temperature being substantially compensated for ...

LEVEL 1 - 105 OF 225 PATENTS

4,753,187

<=2> GET 1st DRAWING SHEET OF 4

Jun. 28, 1988

Individual submarine diving equipment

INVENTOR: Galimand, Patrice, Paris, France

tightness and passage of electric connection between the container and the motor are provided. ... [\*19]

[\*11] 11. The device of claim 1, comprising a control for the motor realized by a moulding-on of a contactor of the deformable thin layer type, said moulding-on having the form of a buckle portion completed by a strip having

pressure connecting means for connection around a hand of the diver, the buckle element being disposable around the diver's ... LEVEL 1 - 106 OF 225 PATENTS

4,729,459

σ GET 1st DRAWING SHEET OF

Mar. 8, 1988

Adjustable damping force type shock absorber

INVENTOR: Inagaki, Mitsuo, Okazaki, Japan Nakano, Hiromichi, Okazaki, Japan Kamiya, Sigeru, Aichi, Japan Ishida, Toshinobu, Okazaki, Japan Sasaya, Hideaki, Ókazaki, Japan Takeda, Kenji, Aichi, Japan

said plunger to that of said other end surface of said sliding ... [\*1] member. [\*2]" 2. An adjustable damping force type shock absorber according to claim 1, wherein said piezo-electric body is a lamination layer type piezo-electric body constituted by laminating in an axial direction a plurality of piezo-electric elements having a piezo-electric effect in which application of a stress in the axial direction ...

piezo-electric body on the basis of the electric signal from the damping force sensor. [\*5] 5. An adjustable damping force type shock absorber according to claim 4, wherein said piezo-electric body is a lamination layer type piezo-electric body constituted by laminating a plurality of piezo-electric elements.

[\*6] 6. An adjustable damping force type shock absorber according to claim 5, wherein said lamination layer type piezo-electric body comprises electrode plates inserted between adjacent piezo-electric elements and forming, respectively, a first electrode by connecting alternate electrode plates in parallel and a ...

LEVEL 1 - 107 OF 225 PATENTS

4,723,601

Feb. 9, 1988

Multi-layer type heat exchanger

INVENTOR: Ohara, Toshio, Kariya, Japan Tsuchiya, Kiyomitsu, Okazaki, Japan Kittaka, Kiyoshi, Aichi, Japan Sudo, Yasuhiro, Okazaki, Japan

Yamauchi, Yoshiyuki, Aichi, Japan Miyata, Yoshio, Nagoya, Japan

What is claimed is:

[\*1] 1. A multi-layer type heat exchanger including:

a row of a plurality of substantially parallel flat tubes each formed by two core plates prepared by press work and sealingly jointed together;

each ...

... [\*7] one outermost core plate to assure that said protecting plate is spaced from said one outermost core plate a distance sufficient to accommodate said further corrugated fin.

8. A multi-layer type heat exchanger including: [\*8] a row of a plurality of substantially parallel flat tubes each formed by two core plates prepared by press work and sealingly jointed together;

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PAGE

4,703,266

LEVEL 1 - 108 OF 225 PATENTS

GET 1st DRAWING SHEET OF <=5>

Oct. 27, 1987

Gradient meter with thin magnetic layer

INVENTOR: Chiron, Guy, Gieres, France
Dumont, Andre , St. Egreve, France

... [\*1] meter to measure the spatial derivatives

delta Hi/ delta j (i = x, y, z; j = x, y, z)

of a magnetic field H utilizing magnetic sonds with a thin layer of the type which comprises: a magnetic layer of cylindrical form of revolution, the magnetic layer having an axis of difficult magnetization parallel to the axis of the cylinder and an axis of easy magnetization that is circular in a plane of the layer in a ... PAGE 120

LEVEL 1 - 109 OF 225 PATENTS

4,702,019

<=2> GET 1st DRAWING SHEET OF 7

\_ .s \_ :

Oct. 27, 1987

Apparatus for cooling high-temperature particles

INVENTOR: Tsuruno, Masayoshi, Funabashi, Japan Horie, Michihiko, Nagareyama, Japan

particles along an outer surface of said conical or pyramidal body is facilitated, and a second packed layer type cooling zone disposed below said first cooling zone for gradually cooling said high-temperature particles while said high-temperature particles which have been rapidly cooled in said first ...  $\dots$  [\*1] said guide tube and is vertically reciprocable so that displacement in the radially outward directions of said high-temperature

LEVEL 1 - 110 OF 225 PATENTS

4,696,548

GET 1st DRAWING SHEET OF <=5>

Sep. 29, 1987

Antiglare mirror for an automobile

INVENTOR: Ueno, Yoshiki, Okazaki, Japan Taguchi, Takasi, Anjo, Japan Hattori, Tadashi, Okazaki, Japan

 $\dots$  [\*1] selected thickness, so that light reflecting from said mirror and passing through said first dielectric layer has desired color characteristics.

[\*2] 2. A mirror arrangement according to claim 1, wherein said liquid crystal layer is of the type whose transparency is reduced when an electric field is applied thereto compared to that when an electric field is not applied thereto.

[\*3] 3. A mirror arrangement according to claim 1, further comprising: second transparent dielectric layer located on the ... LEVEL 1 - 111 OF 225 PATENTS

4,686,159

ന GET 1st DRAWING SHEET OF

Aug. 11, 1987

Laminated layer type fuel cell

INVENTOR: Miyoshi, Hideaki, Kobe, Japan

What is claimed is:

- [\*1] 1. A laminated layer type fuel cell for converting electrochemical reaction of fuel and oxidizer into electric power, said fuel cell comprising a plurality of gas separation plates, each having rectilinear and zigzag portions of fuel and oxidizer ...
- ... [\*1] electrolyte matrix, and an oxidizer electrode whereby partial pressures of reaction gases produced by the electrochemical reaction are equalized throughout the cell to equalize cell reactions and temperature distribution in the cell.
- 2. A laminated layer type fuel cell as claimed in claim 1 wherein the rectilinear and zigzag portions of fuel and oxidizer channels have a length ratio of 1:1 and the zigzag portions are alternately disposed at upstream and downstream sides of the reaction gases.
- 3. A laminated layer type fuel cell as claimed in claim 1 wherein the rectilinear and zigzag portions of fuel and oxidizer channels have a length ratio of 2:1 to 4:1 and the zigzag portions are sequentially displaced with respect to each channel.

LEVEL 1 - 112 OF 225 PATENTS

4,673,591

Jun. 16, 1987

Production of layer-type magnetic recording media

INVENTOR: Lehner, August, Roedersheim-Gronau, Federal Republic of Germany Heil, Guenter, Ludwigshafen, Federal Republic of Germany Lenz, Werner, Bad Durkheim, Federal Republic of Germany Balz, Werner, Limburgerhof, Federal Republic of Germany Kohl, Albert, Laumersheim, Federal Republic of Germany Schornick, Gunnar, Neuleiningen, Federal Republic of Germany

We claim:

[\*1] 1. A process for the production of a layer-type magnetic recording medium by dispersing a finely divided magnetically anisotropic material in a binder which consists of not less than 30% of a radiation-curable aqueous binder dispersion, applying the ...

LEVEL 1 - 113 OF 225 PATENTS

4,671,969

Jun. 9, 1987

Production of layer-type magnetic recording media

INVENTOR: Lehner, August, Roedersheim-Gronau, Federal Republic of Germany Balz, Werner, Limburgerhof, Federal Republic of Germany Lenz, Werner, Bad Durkheim, Federal Republic of Germany

123

Kohl, Albert, Laumersheim, Federal Republic of Germany Heil, Guenter, Ludwigshafen, Federal Republic of Germany

We claim:

binder which consists of not less than 40% of a radiation-curable aqueous binder [\*1] 1. A process for the production of a layer-type magnetic recording medium by dispersing a finely divided magnetically anisotropic material in a dispersion, applying the ...

LEVEL 1 - 114 OF 225 PATENTS

-

4,671,255

<=2> GET 1st DRAWING SHEET OF 2

Jun. 9, 1987

Tissue expander with self-contained injection reservoir and reinforcing insert

reinforcing insert

INVENTOR: Dubrul, William R., Santa Barbara, California Heyler, III, Charles J., Thousand Oaks, California claim 10 wherein the plastic resin embedding the magnetically detectable material is further embedded in a silicone elastomer ... [\*11]

[\*12] 12. A tissue expander device for surgical implantation beneath the skin and the subcutaneous layer of the type which is expanded after implantation by periodic injection of a liquid, such as saline into the expander device, the tissue expander comprising:

a thin expandable biocompatible cast silicone elastomer envelope forming an expandable ...

- tissue expander of claim 15 wherein the magnet in the injection reservoir is embedded in vapor barrier material. ... [\*16]
- skin and the subcutaneous layer of the type which is expanded after implantation by periodic injection of liquid into the expander device, the tissue expander having an expandable blocompatible envelope forming an expandable fluid-tight chamber configured to include an apex and an injection reservoir fully ...
- reservoir whereby location of the injection reservoir can be ascertained by external means for locating the magnet. [\*25]
- skin and the subcutaneous layer of the type which is expanded after implantation by periodic injection of a liquid into the expander device, the tissue expander having an expandable biocompatible envelope forming an expandable fluid-tight 23. In a tissue expander used for surgical implantation beneath the chamber configured to include an apex and an injection reservoir ... [\*23]

PAGE 125

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LEVEL 1 - 115 OF 225 PATENTS

4,667,209

<=2> GET 1st DRAWING SHEET OF 3

May 19, 1987

Image recording apparatus

INVENTOR: Hakamada, Isao, Yokohama, Japan Matsuoka, Kazuhiko, Yokohama, Japan

We claim:

[\*1] 1. An image recording apparatus comprising:

a multi-layer type photosensitive medium; and

means for recording images by scanning said photosensitive medium with a laser beam, said means including a semiconductor laser which produces multimode oscillations, said laser ...

LEVEL 1 - 116 OF 225 PATENTS

4,664,857

May 12, 1987

Process for preparing a hydrogel

INVENTOR: Nambu, Masao, Yokohama, Japan

a water-insoluble hydrogel having a water content of 20 to 92% by  $\dots$  [\*1] weight. [\*2] 2. A process according to claim 1, in which clay minerals of a laminated structure having a three-layer type (2:1 type) composite layer as a basic unit are suspended in said aqueous polyvinyl alcohol solution in an amount of not more than five times by weight the amount of said ...

PAGE 128

LEVEL 1 - 117 OF 225 PATENTS

4,664,843

May 12, 1987

Mixed metal layered hydroxide-clay adducts as thickeners for water and other hydrophylic fluids

INVENTOR: Burba, III, John L., Angelton, Texas
Barnes, Audrey L., Lake Jackson, Texas

vermiculite, chlorite, attapulgite, sepiolite, palygorskite, and Fuller's earth. [\*16]

[\*17] 17. The composition of claim 1 wherein [tne] the mineral clay is at least one of the classes consisting of amorphous clays of the allophane group and crystalline clays of the 2-layer type, 3-layer type, expanding type, non-expanding type, elongate type, regular mixed layer type, and chain structure

[\*18] 18. The composition of claim 1 wherein the mineral clay is bentonite.

[\*19] 19. The composition of claim 1 wherein the mineral clay is beneficiated bentonite.

20. The composition of claim 1 wherein the weight ratio of layered [\*20]

LEVEL 1 - 118 OF 225 PATENTS

4,659,401

<=2> GET 1st DRAWING SHEET OF 2

Apr. 21, 1987

Growth of epitaxial films by plasma enchanced chemical vapor deposition (PE-CVD)

INVENTOR: Reif, L. Rafael, Newton, Massachusetts Fonstad, Jr., Clifton G., Arlington, Massachusetts ... [\*11] establishing a steady state flow of said first set of gaseous reactants in said chamber, said reactants having a concentration of atoms of a first type conductivity, such as to produce a layer of that type conductivity when deposited;

(c) after steady state flow is achieved heating said substrate to a temperature high enough to obtain epitaxial deposition from the first set of reactants when a decomposition reaction occurs ... LEVEL 1 - 119 OF 225 PATENTS

4,644,335

<=2> GET 1st DRAWING SHEET OF 4

Feb. 17, 1987

Apparatus and method for monitoring drill bit condition and depth of drilling

INVENTOR: Wen, Sheree H., Mohegan Lake, New York

number of occurrences of the waveform representing the acoustic ... [\*1] number of occurrences of signature for each type of layer; and means for stopping the drilling operation upon reaching a predetermined count of waveform occurrences for a particular layer type.

- sequence of waveforms stored in said computer corresponding to the multilayered [\*2] 2. The apparatus of claim 1 wherein said control means includes computer for comparing the detected sequence of waveforms to a reference
- type of layer for each signal; and [9\*] ...

bit upon reaching a predetermined count of waveform occurrences for a particular layer type for each drill bit. means for separately stopping the drilling operation of at least one drill

- 7. The apparatus of claim 1 or 6 further including a filter means for filtering out low and high frequency noise.
- [\*8] "8. In a multiple ...

proper drill bit drilling having each type of layer, for counting the number of occurrences of the waveform representing the acoustic signature for each layer type for each of said drill bits, and for detecting when the acoustic signature of at least one of said output signals is different from the acoustic signature of at least one of said output signals is different from the acoustic signature of said reference signals is different from the acoustic signature.

bit upon detection of an improper drill bit condition; and [8\*] ... means for stopping the drilling operation of at least one drill bit upon reaching a predetermined count of waveforms occurrences of a particular layer type for one or more drill bits.

[\*9] 9. A method for drilling to a predetermined depth of a multilayer workpiece comprising:

acoustically detecting drill bit vibrations as the drill bit ...

layer; [6\*] ...

Pat. No. 4644335, \*9

counting the number of occurrences of the waveform representing the acoustic signatures for each type of layer; and

stopping the drilling operation upon reaching a predetermined count of waveform occurrences for a particular layer type.

<code>[\*10]</code> 10. The method of claim 9 further including the steps of producing an output signal having a sequence of waveforms representing the acoustic signatures corresponding to the sequence of layers, comparing the sequence of waveforms to a reference sequence and ...

least one of said output signals and said reference signal; and

predetermined count of waveform occurrences for a particular layer type for one or more drill bits. stopping the drilling operation of at least one drill bit upon reaching a

PAGE

LEVEL 1 - 120 OF 225 PATENTS

4,629,632

Dec. 16, 1986

Production of magnetic recording media

INVENTOR: Balz, Werner, Limburgerhof, Federal Republic of Germany Kovacs, Jenoe, Hessheim, Federal Republic of Germany Lechner, Hilmar, Frankenthal, Federal Republic of Germany Schaefer, Dieter, Lindenberg, Federal Republic of Germany Buethe, Ingolf, Boehl-Iggelheim, Federal Republic of Germany

We claim:

medium by applying a dispersion of a magnetically anisotropic material in a binder solution onto a flexible plastic base provided with an adhesion-promoting intermediate layer and then solidifying the ...

LEVEL 1 - 121 OF 225 PATENTS 1. A process for the production of a layer-type magnetic recording [\*1]

4,617,423

9 GET 1st DRAWING SHEET OF

Oct. 14, 1986

Data communication system

INVENTOR: Dickerson, James W., Plano, Texas Smith, III, William N., Carrollton, Texas

What is claimed is:

[\*1] 1. A network multiple physical layer interface connected to a communcations network of a first physical layer type and a second physical layer type, each layer type including a send channel and a receive channel, said interface comprising:

a first circuit means for receiving data from said first physical layer send channel and receive channel and ... LEVEL 1 - 122 OF 225 PATENTS

134

PAGE

4,614,185

GET 1st DRAWING SHEET OF 1

Sep. 30, 1986

Piston engine having a phosphatized cylinder wall

INVENTOR: Fox, Richard C., Mobile, Alabama

... [\*4] 1, wherein said integral layer of crystalline phosphate is characterized by resistance to wear by said piston ring during operation of said

[\*5] 5. The invention according to claim 1, wherein said integral layer is of the type assisting the seating of said piston ring in said cylinder.

 $[\star 6]$  6. The invention according to claim 1, wherein said cylinder head is an aluminum cylinder head.

[\*7] % 7. The invention according to claim 6, wherein said ... GE 135

LEVEL 1 - 123 OF 225 PATENTS

4,611,114

~ <=2> GET 1st DRAWING SHEET OF

Sep. 9, 1986

Photoelectric detection structure having substrate with controlled properties

INVENTOR: Dolizy, Pierre, Ris-Orangis, France Groliere, Francoise, Nogent-sur-Marne, France Maniguet, Francois, Fontenay-Tresigny, France

... [\*8] according to claim 7, wherein said tri-alkaline material is SbNa2K, Cs.

[\*9] 9. A photoelectric detection structure according to claim 7, wherein said photosensitive layer has a thickness corresponding to a photoelectric layer of the type S20 or S25.

 $[*10]\ 10.$  A photoelectric detection structure according to claim 2, wherein said photosensitive layer is a bi-alkaline photosensitive material.

A

LEVEL 1 - 124 OF 225 PATENTS

4,604,673

Aug. 5, 1986

Distribution transformer with surge protection device

INVENTOR: Schoendube, Charles W., Hickory, North Carolina

What I claim as new is:

[\*1] 1. A distribution-type single-phase transformer having a surge protection arrangement comprising;

(a) a layer-type high voltage winding having two terminals, one being a high voltage terminal for connection to a high voltage line,

(b) a divided low voltage winding comprising two ... LEVEL 1 - 125 OF 225 PATENTS

4,587,720

<=2> GET 1st DRAWING SHEET OF 1

May 13, 1986

Process for the manufacture of a self-aligned thin-film transistor

INVENTOR: Chenevas-Paule, Andre , Grenoble, France Diem, Bennard, Meylan, France

... [\*2] wavelength of the order of 600 nanometers.

[\*3], at 3. A process according to claim 1, wherein step (h) comprises:

depositing a layer of n + type amorphous silicon on the entire structure;

depositing a conducting layer on the type n + silicon layer;

eliminating the regions respectively of the conducting layer and of the type n + silicon layer situated in line with said grid; and

making the electrodes of the source and of the drain in said conducting

[\*4] 4. A process according to claim 1, wherein said insulating layer is PAGE 138

137 PAGE

LEVEL 1 - 126 OF 225 PATENTS

4,584,553

Apr. 22, 1986

Coated layer type resistor device

INVENTOR: Tokura, Norihito, Nukata, Japan Kawai, Hisasi, Toyohashi, Japan

We claim:

[\*1] 1. A coated layer type resistor device comprising:

an insulator substrate;

a first resistor element formed on said insulator substrate and consisting of a resistor layer and end conductor electrodes at the ends of said resistor layer; and

... p

... [⋪] conductor electrodes and the adjacent intermediate conductor in said second resistor element being equal to the distance between end conductor electrodes in said first resistor element.

2. A coated layer type resistor device according to claim 1, wherein: [\*3]

said end conductor electrodes and said intermediate conductors are formed by printing on said insulator substrate;

on said insulator substrate having said formed end conductor electrodes and intermediate conductors, a resistor layer is formed by printing; and

said formed resistor layer is in contact with said formed end conductor electrodes and intermediate conductors.

[\*3] 3. A coated layer type resistor device according to claim 1, wherein a bridge circuit is constituted by said first and second resistor elements. PAGE 139

LEVEL 1 - 127 OF 225 PATENTS

4,576,116

GET 1st DRAWING SHEET OF 1

Mar. 18, 1986

Collapsible house for cats

INVENTOR: Binkert, Gerald A., 308 Gould Ave. SE., Bemidji, Minnesota 56601

- A collapsible A-frame house providing a common site for a cat to rest, exercise and play, comprising
- (a) a roof formed of a single unitary continuous flexible layer-type material having a backside and a cushiony outward face, said roof including an elongate peak and two roof panels integral with the peak and depending downwardly and outwardly from the peak, each said roof panel having a bottom edge, the outward face of the flexible layer-type material being the outward face of the roof and being adapted to withstand cat clawing, the peak of the roof being adapted for flexing in a hinge-like manner to permit inward movement of the roof ...
- ... [\*1] another when the house is to be collapsed;
- (b) a stiffening means on the backside of each roof panel for supporting the Same:
- material having a bottom side and a cushiony top side, said floor panel having opposing edges thereof affixed to the bottom edges of the roof panels to limit the separation distance between said bottom edges, the floor panel being adapted for folding along its longitudinal center line generally parallel to its opposing edges, and the flexible layer-type material adjacent the junction of the bottom edges of the roof panels and the opposing edges of the floor panel being such as to serve a hinge function for allowing the floor panel to fold as (c) a floor panel formed of a single unitary continuous flexible layer-type the roof panels ...
- A collapsible A-frame house providing a common site for a cat to rest, exercise and play, comprising
- (a) a roof formed of a single unitary continuous flexible layer-type material having a backside and a cushiony outward face, said roof including an elongate peak and two roof panels integral with the peak and depending downwardly and outwardly from the peak, each said roof panel having a bottom edge, the outward face of the flexible layer-type material being the outward face of the roof and being adapted to withstand cat clawing, the peak of the roof being adapted for flexing in a hinge-like manner to permit inward movement of the roof...
- ... [\*10] another when the house is to be collapsed;
- (b) a stiffening means on the backside of each roof panel for supporting the same; and
- material having a bottom side and a cushiony top side, said floor panel having opposing edges thereof affixed by staples to the bottom edges of the roof (c) a floor panel formed of a single unitary continuous flexible layer-type

Pat. No. 4576116, \*10

panels to limit the separation distance between said bottom edges, the floor panel being adapted for folding along its longitudinal center line generally parallel to its opposing edges, and the flexible layer-type material adjacent the junction of the bottom edges of the roof panels and the opposing edges of

the floor panel being such as to serve a hinge function for allowing the floor panel to fold as the roof panels ... LEVEL 1 - 128 OF 225 PATENTS

141

PAGE

4,566,460

GET 1st DRAWING SHEET OF 13

Jan. 28, 1986

Measuring method and apparatus for non-linear parameter of acoustic medium and its application

INVENTOR: Sato, Takuso, Tokyo, Japan Ichida, Nobuyuki, Machida, Japan Miwa, Hirohide, Kawasaki, Japan

 $\dots$  [\*20] reception characteristics sufficient to cover both receiving said probing wave and transmitting said pumping wave.

[\*21]  $^{(*21)}$  An apparatus according to claim 10, wherein said second and said third transducer comprise a layer type transducer, having a front layer as said second transducer, and a back layer as said third transducer.

22. An apparatus according to claim 6, wherein said third means comprises: [\*22]

phase ...

LEVEL 1 - 129 OF 225 PATENTS

4,560,419

3 GET 1st DRAWING SHEET OF <=5>

Dec. 24, 1985

Method of making polysilicon resistors with a low thermal activation energy

INVENTOR: Bourassa, Ronald R., Colorado Springs, Colorado Butler, Douglas B., Colorado Springs, Colorado

... [\*11] 11. The method of claim 7 including establishing said first, second and third poly regions to form back-to-back polysilicon diodes.

[\*12] 12. The method of claim 8 including doping a poly layer with the type of impurity for said first region of poly, then defining the poly which is to act as the resistor, the selectively doping said second and third regions of poly with the other type of impurity.

13. The method of claim 12 wherein the ... LEVEL 1 - 130 OF 225 PATENTS [\*13]

4,547,784

:=2> GET 1st DRAWING SHEET OF 6

Oct. 15, 1985

Thermal recording system and method

INVENTOR: Erlichman, Irving, Wayland, Massachusetts
Hausslein, Robert W., Lexington, Massachusetts

layer of the type wherein recorded dot size increases with increased amounts of density on a transparency type thermally sensitive recording medium having transparent support layer and a transparent thermally sensitive recording thermal energy applied to form a dot, said recording system comprising:

means for supporting such a transparency type of recording medium;

means.

LEVEL 1 - 131 OF 225 PATENTS

4,525,223

<=2> GET 1st DRAWING SHEET OF 16

Jun. 25, 1985

Method of manufacturing a thin ribbon wafer of semiconductor material

INVENTOR: Tsuya, Noboru, 1-38, Kashiwagi 2-Chome, Sendai, Japan Arai, Keņichi, Sendai, Japan ... [\*18] moving direction of the cooling substrate and at least two jet flows of semiconductor and gaseous or molten material including the same semiconductor are simultaneously ejected through the holes so as to form a thin ribbon of multiple-layer type.

heat-treated at a temperature within the range from 5000 C. to a melting point for a time of 0.1 ... [\*19] 19. A method as defined in claim 1, wherein the thin ribbon is

LEVEL 1 - 132 OF 225 PATENTS

4,523,906

<=2> GET 1st DRAWING SHEET OF 8

Jun. 18, 1985

Device for drying gypsum

4GE 144

INVENTOR: Petrovic, Vladan, Essen, Federal Republic of Germany

- plurality of heat-retaining solid particles into said heater; means for storing agypsum to be dried; a moving-layer-type drier in the form of an upright stationary container having an inlet at its top, an outlet at its bottom and a plurality of superposed funnel-like means arranged between the inlet and the outlet; means for conveying an amount of hot ...
- ... [\*6] cold, wet gypsum, comprising a heater; means for feeding a plurality of heat-retaining solid particles into said heater; means for storing gypsum to be dried; a moving-layer-type drier in the form of an upright stationary container having an inlet at its top, an outlet at its bottom and a plurality of superposed funnel-like means arranged between the inlet and outlet; means for intermixing the amount of hot ...

4,513,016

Apr. 23, 1985

No-stir dry mix with pudding nuggets for cake with

INVENTOR: Blake, Jon R., 6901 Regent Ave N., Brooklyn Center, Minnesota 55429 Knutson, Richard K., 6948 Valley View Rd., Corcoran, Minnesota 55340 VanHulle, Glenn J., 7608 Major Ave. N., Brooklyn Park, Minnesota 55443 discontinuous pudding phase

... [\*14] length.

[\*15] 15. The dry mix of claim 14 wherein the weight ratio of sugar to granules in the matrix ranges from about 1:5 to 1:6.

[\*16] 16. A method for preparing a finished cake of a layer type having after baking a discontinuous pudding phase, in the finished baked cake consisting essentially of the steps of:

A. providing a dry mix for cakes, said dry mix comprising

I. from ...

LEVEL 1 - 134 OF 225 PATENTS

4,510,443

4 GET 1st DRAWING SHEET OF <=5>

Apr. 9, 1985

Voltage measuring device

INVENTOR: Inaba, Ritsuo, Hirakata, Japan Wasa, Kiyotaka, Nara, Japan

What is claimed is:

 $[\star 1]$  1. A voltage measuring device for receiving and measuring a voltage to be measured and for providing an output signal corresponding thereto, said device comprising: a first medium of the double layer type for propagating surface acoustic waves, said first medium comprising a piezoelectric thin film which is laminated on a substrate comprising a non-piezoelectric material;

a first transducer means ...

AGE 148

LEVEL 1 - 135 OF 225 PATENTS

4,506,004

<=2> GET 1st DRAWING SHEET OF 3

Mar. 19, 1985

Printed wiring board

INVENTOR: Sullivan, Donald F., 115 Cambridge Rd., King of Prussia, Pennsylvania 19406

.. [\*5] substrate, and

developing the photo images by washing out the unhardened photopolymer, whereby removal of the unhardened liquid polymer layer in contact with the substrate is simple and complete.

[\*6] 6. The process of claim 5 wherein the laminated photopolymer layers are of the type that are hardened by exposure to the radiation.

[\*7] 7. The photo process of claim 6 including the step of partly curing the liquid photopolymer second layer by exposure to radiation before lamination.

[\*8] 8. The photo process of ... LEVEL 1 - 136 OF 225 PATENTS

4,501,303

<=2> GET 1st DRAWING SHEET OF 2

Feb. 26, 1985

Forming fabric

INVENTOR: Osterberg, Lars B., Halmstad, Sweden

What I claim is:

<code>[\*1]</code> 1. An improved double-layer type of forming fabric for use in papermaking, cellulose and similar machines, said forming fabric consisting of two integral weaves, each one of said weaves comprising its separate sets of respective warp ...

LEVEL 1 - 137 OF 225 PATENTS

4,494,826

<=2> GET 1st DRAWING SHEET OF 2

Jan. 22, 1985

ध्र

INVENTOR: Smith, James L., 426 High School Dr., Grand Prairie, Texas 75050

 $\dots$  [\*2] set forth in claim 1 wherein said grille structure layer is inside said photoconductive layer.

[\*3] 3. A device as set forth in claim 1 or 2 wherein said photoconductor

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Subject: LEXIS(R)/NEXIS(R) Print Request Job 68990, 3 of

layer is of the type which requires high electric field for significant photoconduction such as CdS powder in plastic.

4. A device as set forth in claim 1 wherein a nonconducting light blocking ...

PAGE

LEVEL 1 - 138 OF 225 PATENTS

4,477,547

Oct. 16, 1984

Method for making complex layer type lithografic printing

1000 E

INVENTOR: Yamada, Jun, Nagaokayo, Japan Suzuki, Shigeyoshi, Nagaokayo, Japan Senga, Takao, Nagaokayo, Japan

What is claimed is:

[\*1] 1. A method for making a complex layer type lithographic printing plate which comprises forming a toner image on an original printing plate having organic electrophotographic photosensitive layer by electrophotographic process, said photosensitive layer being a complex layer type photosensitive layer which comprises a charge carrier generating layer comprising a charge carrier generating substance and a binder mainly composed of a polyamide resin soluble in alcohol solvent and ...  $\dots$  [\*] solution mainly composed of alcohol solvent and/or alkali solvent and thereafter treating non-image area other than the toner image area with the etching solution.

[\*2] 2. A method for making a complex layer type lithographic printing plate according to claim l, wherein the polyamide resin of the binder for the charge carrier generating layer is copolymer nylon.

- [\*3] 3. A method for making a complex layer type lithographic printing plate according to claim 1 wherein the high molecular substance of the binder for the charge carrier generating layer is one having acid anhydride group, carboxylic acid group, sulfonic acid group or sulfonimide group.
- [\*4] 4. A method for making a complex layer type lithographic printing plate according to claim 1 wherein the charge carrier generating substance is organic pigment or sensitizing dye.
- an plate according to claim 1 wherein the charge carrier transport substance is aromatic tertiary diamino compound, an aromatic tertiary diamino compound, an aromatic tertiary triamino compound, a condensate or a heterocyclic compound. 5. A method for making a complex layer type lithographic printing
- [\*6] 6. A method for making a complex layer type lithographic printing plate according to claim 1 wherein the high molecular substance contained in the binder for the charge carrier transfer layer is one having acid anhydride group, carboxylic acid group, sulfonic acid group or sulfonimide group or a phenolic
- [\*7] 7. An original printing plate having organic electrophotographic photosensitive layer on a support for complex layer type lithographic printing plate on which a toner image is formed, said photosensitive layer comprising a charge carrier generating layer comprising a and a binder mainly ...

Pat. No. 4477547, \*7

- ... [\*7] a high molecular substance having a group soluble in an etching solution mainly composed of alcohol solvent and/or alkali solvent.
- [\*8] 8. A printing method which comprises carrying out printing with the complex layer type\_lithographic printing plate made by the method of claim 1.

LEVEL 1 - 139 OF 225 PATENTS

4,470,624

<=2> GET 1st DRAWING SHEET OF 2

Sep. 4, 1984

Integrated circuit for a controllable frequency oscillator

INVENTOR: Leuenberger, Claude-Eric, Chezard, Switzerland

- ... [\*1] said insulating layer, said chip having an oxide layer, said insulating layer being a portion of said oxide layer.
- [\*2] 2. The chip of claim 1, wherein said first region is a bulk layer of the type of conductivity opposite to said one type of conductivity, said bulk layer being formed in said semiconductor substrate, and said second region is

4GE 152

diffusion portion of said one type of conductivity ... LEVEL 1 - 140 OF 225 PATENTS

4,451,843

GET 1st DRAWING SHEET OF

May 29, 1984

Bipolar transistor with a plurality of parallelly connected base-collector junctions formed by plastic deformation of

the crystal lattice

INVENTOR: Dahlberg, Reinhard, Flein, Federal Republic of Germany

semiconductor plate, disc or chip have a highly doped surface layer with the type of conductivity of the base region; both semiconductor discs are assembled, by mechanical pressure, so that the ridges of the structured faces of both plates, discs or chips cross and touch; and the surfaces of said ridges which  $\dots$  [\*1] emitter region on its opposite main face so as to form an emitter-base p/n junction; the side surfaces of the ridges in the other are in contact are connected

ed ... LEVEL 1 - 141 OF 225 PATENTS

4,427,607

9 GET 1st DRAWING SHEET OF

Jan. 24, 1984

Device in an evaporative cooler

INVENTOR: Korsell, Lars E. R., Stockholm, Sweden

[\*4] telescopically received only in the corresponding top openings of a similar contact body immediately therebelow. [\*5] 5. In an evaporative cooler including, a casing, at least one contact body of the multi-layer type located in said casing and formed with channels existing between the layers and which all are passed by air, and means for supplying water to selected channels in said body from above the body, the improvement comprising ...

LEVEL 1 - 142 OF 225 PATENTS

4,422,627

GET 1st DRAWING SHEET OF

Dec. 27, 1983

Endless spring, such as ringspring

INVENTOR: Schmidt, Helmut, Munich, Neubiberg, Federal Republic of Germany Ramm, Ulrich, Neubiberg, Federal Republic of Germany Schroeder, Alexander, Ottobrunn, Federal Republic of Germany

- made of fiber compound materials and extending in parallel to said frame plane, said layers compound materials and extending in parallel to said frame plane, said layers comprising a first layer type (2) in which fiber bundles are wound so that all the fibers extend unidirectionally in said first layer type and a second layer type (3, 4), each said second layer type comprising at least two plies (5, 6) in which the fibers extend in cross-over relationship relative to each other, said first layer type and said second layer type being arranged in alternate succession relative to each other.
- layer type forms inner layers interconnected by a second type layer between two comprises one layer more than said first layer type so that the second layer type type forms outer surfaces parallel to said frame plane, and so that said first neighboring first type layers.
- [\*3] 3. The endless spring of claim 1, wherein said plies (5, 6) of said second layer type (3, 4) comprise at least one first ply (5) with fibers extending in parallel and at 900 relative to said main load application direction, and at least one second ply (6) with fibers extending in a +/- 450 cross-over relationship relative to said main load application direction.
- [\*4] 4. The endless spring of claim 3, wherein each of said second layer type (3, 4) comprises said first and second plies (5, 6), wherein a second layer type (3) located internally of the spring between two first layer types (2) comprises at least two first plies (5) and a second ply (6) located between said two first plies (5), and wherein a second layer type (4) located externally of the spring comprises at least one first ply (5) and at least one second ply (6) located on the outside of the spring.
- [\*5] 5. The spring of claim 1, wherein each of said first layer type (2) comprises at least two fiber bundles.
- [\*6] 6. The spring of claim 1, wherein said plies of said second layer type are made of fibers of different materials.
- [\*7] 7. The spring of claim 1, wherein said fiber bundles of said first layer type (2) are made of fibers of different materials.
- [\*8] 8. The spring of claim 1, wherein said fiber bundles of said first layer type (2) and said plies of said second layer type (3, 4) are made of fibers of different materials.

Pat. No. 4422627, \*8

[\*9] 9. The spring of claim 1, further comprising adhesive layers (7) operatively interposed between said first and second layer types.

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[\*10] 10. The spring of claim 1, further comprising spring attachment means (8) operatively secured to said endless spring for applying a load to the spring, and wear resistant plate means (9) ...

 $\dots$  [\*10] reducing the wear imposed by the atrachment means on the spring

[\*11] 11. The spring of claim 10, wherein said plate means (9) are made of spring steel.

[\*12] 12. The spring of claim 1, wherein said first and second layer types (2, 3, 4) form an endless loop having two straight legs extending in parallel to each other and two curved end portions operatively interconnecting said straight

[\*13] 13. The spring of ... LEVEL 1 - 143 OF 225 PATENTS

4,419,310

=2> GET 1st DRAWING SHEET OF 2

Dec. 6, 1983

SrTi03 barrier layer capacitor

INVENTOR: Burn, Ian, Williamstown, Massachusetts Neirman, Stephen M., Williamstown, Massachusetts

What is caimed is:

[\*1] 1. A method for making an intergranular barrier layer type capacitor without heating in a reduced atmosphere comprising:

(a) preparing a ceramic start mixture consisting essentially of strontium, titanium and strontium-titanate donor compounds, said donors being ...

LEVEL 1 - 144 OF 225 PATENTS

4,414,059

<=2> GET 1st DRAWING SHEET OF 3

Nov. 8, 1983

Far UV patterning of resist materials

INVENTOR: Blum, Samuel E., White Plains, New York Brown, Karen H., Yorktown Heights, New York Srinivasan, Rangaswamy, Ossining, New York

final step thereof includes the treatment of said exposed portions ... [\*7] final step thereof includes the treatment of said export said substrate through said patterned resist layer to modify the characteristics of said exposed portions.

[\*8] 8. A method for patterning resist layers of the type used in lithography processes, comprising the steps of:

depositing a layer of said resist on a substrate, and

irradiating selected areas of said resist layer with ultraviolet radiation having wävelengths less than 220 nm and an ... LEVEL 1 - 145 OF 225 PATENTS

4,411,539

GET 1st DRAWING SHEET OF 7

Oct. 25, 1983

Print element with plural type layers of varying thickness

INVENTOR: Iwata, Nobuo, Sagamihara, Japan Hasegawa, Takashi, Hiratsuka, Japan

... [\*1] by the hammer, and the vertical dimension of the character carried by the type member; such variance in thicknesses eliminating ghost printing caused by the undesired touching of the paper by the type member adjacent in layer to the type member struck by the member. [\*2] 2. A print element as claimed in claim 1, wherein the print element is formed in a form of a disc type print wheel including ... LEVEL 1 - 146 OF 225 PATENTS

4,405,533

4 GET 1st DRAWING SHEET OF

Sep. 20, 1983

Supply device for use with evaporative contact bodies

INVENTOR: Norback, Per, Lidingo, Sweden Eriksson, Borje, Sigtuna, Sweden

We claim:

[\*1] 1. A supply device for use in a evaporative contact body of the multi-layer type having gaps between the layers of the body to which water is supplied from above and air is supplied from below, said device comprising water supply pipes and means for forming jets of water and directing said water jets ...

161 PAGE

LEVEL 1 - 147 OF 225 PATENTS

4,403,236

Sep. 6, 1983

Boundary layer type semiconducting ceramic capacitors with high capacitance

INVENTOR: Mandai, Haruhumi, Nagaokakyo, Japan Nishimura, Kunitaro, Youkaichi, Japan Yamaguchi, Masami, Nagaokakyo, Japan

What we claim is:

Ø [\*1] 1. A boundary layer type semiconducting ceramic capacitor comprising semiconducting ceramic body in which grain boundaries on crystal grans of the semiconducting ceramic body are insulated, characterized in that said semiconducting ceramic body has a composition ...

... [\*1] consisting of Mn, Bi, Cu, Pb, B and Si, and that the maximum crystal grain present in the semiconducting ceramic body has a grain size ranging from 100 mu to 250 mu .

2. The boundary layer type semiconducting ceramic capacitor according to claim 1 wherein said composition contains 0.02 to 0.2 mole % of Mn [\*3] 3. The boundary layer type semiconducting ceramic capacitor according to claim 2 wherein Mn is present in at least one of the grain boundaries and the crystal grains.

to claim 1 wherein said composition further contains at least one of 0.05 to 0.5 mole % of Si02 and 0.02 to 0.2 mole % of Al203. 4. The boundary layer type semiconducting ceramic capacitor according

[\*5] 5. The boundary layer type semiconducting ceramic capacitor according to claim 2 wherein said composition further contains at least one of 0.05 to 0.5 mole % of Si02 and 0.02 to 0.2 mole % of Al203.

6. The boundary layer type semiconducting ceramic capacitor according to claim 2 wherein Mn is present in the crystal grains. [\*7] 7. The boundary layer type semiconducting ceramic capacitor according to claim 2 wherein Mn is present in the grain boundaries of the crystal grains, and wherein said grain boundaries of the crystal grains are insulated by Mn and at least one other insulating agent. [\*8] 8. The boundary layer type semiconducting ceramic capacitor according to claim 1 in which the amount of the mair component is 98.1 to 99.88 mole %.

[\*9] 9. The boundary layer type semiconducting ceramic capacitor according to claim 8 wherein Mn is present at at least one of the grain boundaries and the crystal" grains. [\*10] 10. The boundary layer type semiconducting ceramic capacitor according to claim 9 wherein Mn is present at the crystal grains. GE 163

Pat. No. 4403236, \*10

[\*11] 11. The boundary layer type semiconducting ceramic capacitor according to claim 10 wherein said composition further includes at least one of 0.05 to 0.5 mole % of SiO2 and 0.02 to 0.2 mole % of Al2O3.

[\*12] 12. The boundary layer type semiconducting ceramic capacitor according to claim 9 wherein said composition further includes at least one of 0.05 to 0.5 mole % of Si02 and 0.02 to 0.2 mole % of Al203.

[\*13] 13. The boundary layer type semiconducting ceramic capacitor according to claim 8 wherein said main component is Sr1 - xBa[x] 103. [\*13]

[\*14] 14. The boundary layer type semiconducting ceramic capacitor according to claim 1 wherein said main component is (Sr1 - xBa[x])Ti03. PAGE 164

LEVEL 1 - 148 OF 225 PATENTS

4,397,886

GET 1st DRAWING SHEET OF 1 <=5>

Aug. 9, 1983

Method for making a ceramic intergranular barrier-layer capacitor

INVENTOR: Neirman, Stephen M., Williamstown, Massachusetts Burn, Ian, Williamstown, Massachusetts

What is claimed is:

[\*1]  $^{11}$  . A method for making a ceramic intergranular barrier layer type capacitor comprising: (a) preparing a ceramic start mixture consisting essentially of strontium, titanium, a strontium-titanate-donor and manganese, said donor being selected from large cations A, small

LEVEL 1 - 149 OF 225 PATENTS

May 31, 1983

Stable silicone-coated release liner for pressure-sensitive adhesive sheets

165

INVENTOR: Campbell, Karen J., Anoka, Minnesota Evans, Jack L., St. Paul, Minnesota ... [\*1] hydrogen in (2) to silicon-bonded ethylenically unsaturated radicals in (1) being from 1:1 to 20:1.

 $[\star 2]$  2. The release liner of claim 1 wherein each side of the base sheet is provided with a silicone layer of the type defined.

3. The release liner of claim 1 wherein R and R<1 > are both methyl

[\*4] 4. The release liner of claim 1 wherein R<2 > is a vinyl radical.

[\*5] 5. The release liner of claim ... LEVEL 1 - 150 OF 225 PATENTS

4,363,997

<=2> GET 1st DRAWING SHEET OF 2

Dec. 14, 1982

Dec.

Fluorescent lamp having reflective layer

INVENTOR: Kodama, Churyo, Ohme, Japan

What is claimed is:

[\*1] 1. A fluorescent lamp of the reflective layer type comprising a glass tube, a first phosphor layer formed on the entire face of the inner wall of said glass tube and a second phosphor layer formed on said first phosphor layer at

... [\*1] m and the average particle size of the phophor constituting said first phosphor layer is smaller than the average particle size of the phosphor constituting said second phosphor layer.

[\*2] 2. A fluorescent lamp of the reflective layer type as set forth in claim 1, wherein the average particle size of the phosphor constituting the first phosphor layer is smaller than 10 mu m.

[\*3] 3. A fluorescent lamp of the reflective layer type as set forth in claim 1, wherein the average particle size of the phosphor constituting the second phosphor layer is smaller than 30 mu m.

[\*4] 4. A fluorescent lamp of the reflective layer type as set forth in claim 1, wherein each of the amounts coated of the phosphors of the first and second phosphor layers is 2 to 4 mg/cm<2> .

[\*5] 5. A fluorescent lamp of the reflective layer type as set forth in claim 1, wherein the reflection angle is in the range of 180oto 240o.

- [\*6] 6. A fluorescent lamp of the reflective layer type as set fort in claim  $5 \mid \cdot \cdot \cdot \cdot \cdot \cdot \cdot$  wherein the reflection angle is 1800.
- [\*7] 7. A fluorescent lamp of the reflective layer type as set forth in claim 1, wherein the phosphor constituting the first phosphor layer has the same light emission spectrum as that of the phosphor constituting the second phosphor layer.
- [\*8] 8. A fluorescent lamp of the reflective layer type as set forth in claim 1, wherein the phosphor constituting the first phosphor layer has a light emission spectrum different from that of the phosphor constituting the second phospher layer.
- [\*9] 9. A fluorescent lamp of the reflective layer type as set forth in claim 1, wherein the glass tube is a straight tube.
- [\*10] 10. A fluorescent lamp of the reflective layer type as set forth in claim 1, wherein the glass tube is a circular or curved tube. 167

LEVEL 1 - 151 OF 225 PATENTS

4,363,769

œ GET 1st DRAWING SHEET OF

Dec. 14, 1982

Method for manufacturing thin and flexible ribbon wafer of semiconductor material and ribbon wafer INVENTOR: Tsuya, Noboru, 1-38, Kashiwagi 2-Chome, Sendai City, Japan Arai, Kenichi, Sendai, Japan

- ... [\*18] extending parallel to a moving direction of the ejected melt so that at least two jet flows of some or different semiconductor material are simultaneously ejected through the holes so as form a thin ribbon wafer or multi-layer type.
- [\*19] 19. A method as defined in claim 1, wherein the raw semiconductor material is mixed with a substance selected from the group consisting of Ge, Si, Se, Te, PbS, InSb, ZnTe, PbSe, ...

LEVEL 1 - 152 OF 225 PATENTS

4,362,597

GET 1st DRAWING SHEET OF 1

÷s it.

Dec. 7, 1982

silicide-on-polysilicon structures for MOS devices Method of fabricating high-conductivity

INVENTOR: Fraser, David B., Berkeley Heights, New Jersey Kinsbron, Eliezer, Highland Park, New Jersey Vratny, Frederick, Berkeley Heights, New Jersey

... [\*1] containing layer on top of said pattern and on said selected regions, the metallic constituent in said layer being selected from the group consisting of titanium, tantalum, molybdenum, tungsten, nickel and cobalt, which metal-containing layer is of the type that, upon sintering, will form a silicide.

lifting off said pattern thereby leaving on said device only the metal-containing layer deposited on said selected regions,

sintering said remaining metal- ...

... [\*4] top of said masking pattern and on said selected surface regions, the metallic constituent in said layer being selected from the group consisting of titanium, tantalum, molybdenum, tungsten, nickel and cobalt, which metal-containing layer is of the type that, upon sintering, will form a

removing said masking pattern from said polysilicon layer thereby lifting off those portions of said metal-containing layer deposited on top of said masking pattern and ... ... [\*5] masking pattern and on said surface regions of said polysilicon layer, the metallic constituent in said layer being selected from the group consisting of titanium, tantalum, molybdenum, tungsten, nickel and cobalt, which metal-containing layer is of the type that, upon sintering, will form a

removing said masking pattern from said intermediate layer thereby lifting off those portions of said metal-containing layer deposited on top of said masking pattern and ...

LEVEL 1 - 153 OF 225 PATENTS

4,362,158

2 GET 1st DRAWING SHEET OF

Dec. 7, 1982

fractions, perfusion solutions, dialysis solutions and Synthetic bag-type container for human blood and its alimentary and chemical liquids in general INVENTOR: Lena, Paolo, Via Castello, 13, 26038 Torre de'Picenardi (Cremona),

... [\*3] two parallel side edges of the container.

[\*4] 4. A container as claimed in claim 1, particularly for containing solutions and liquids in general, wherein said initial film sheet is of the single-layer type, and is constituted by polyethylene-butyl rubber copolymer, polyethylene or polypropylene.

[\*5] 5. A container as claimed in claim 1, wherein the initial film sheet has a thickness of 80-150 microns.

\*6| 6.

LEVEL 1 - 154 OF 225 PATENTS

PAGE 170

4,360,819

<=2> GET 1st DRAWING SHEET OF 4

Nov. 23, 1982

Thermal recording apparatus

INVENTOR: Saito, Tamio, Oume, Japan Fukumoto, Yoshikatsu, Hamura, Japan Tagaya, Kiyomi, Oume, Japan  $\dots$  [\*5] respective switching group and capable of limiting the switching operation of said respective switching group.

[\*6] 6. A thermal recording apparatus according to claim 1, wherein said capacitor is an electrolytic capacitor of an electric double layer type construction. [\*7] 7. A thermal recording apparatus according to claim 1, wherein said detecting means comprises two serially connected resistors in parallel with said capacitor; and a comparator means, coupled to ...

 $\dots$  [\*11] signals stored in said memory when the detected terminal voltage is less than the predetermined voltage.

[\*12] 12. A thermal recording apparatus according to claim 10, wherein said capacitor is an electrolytic capacitor of electric double layer type construction.

LEVEL 1 - 155 OF 225 PATENTS

4,352,116

<=2> GET 1st DRAWING SHEET OF 7

Sep. 28, 1982

## Solid state electro-optical devices on a semi-insulating substrate

INVENTOR® Yariv, Amnon, San Marino, California Margalit, Shlomo, Pasadena, California Lee, Chien-Ping, Pasadena, California

۵ ... [\*15] matching parameters, with adjacent semi-conductor layers having different combinations of constituent elements and being of either of the N or type, each of said layers including a region which is doped to a type opposite the layers's type whereby a PN junction is formed in the second layer; and matching parameters, with adjacent semi-conductor layers having

172 PAGE a first and second contacts on the top surface of the top third layer, said first contact being on the surface which is not doped and the second contact on

LEVEL 1 - 156 OF 225 PATENTS

4,341,686

Jul. 27, 1982

Adhesive products and a process for their use in polyurethanes

INVENTOR: Chakrabarti, Sarbananda, Ludwigshafen, Federal Republic of Germany Hutchison, John, Wachenheim, Federal Republic of Germany Volkert, Otto, Weisenheim, Federal Republic of Germany

... [\*6] by weight of a solvent mixture of cyclohexanone/methylene chloride in a weight ratio of approximately 50:50.

[\*7] 7. A process for improving the adhesiveness of cellular or noncellular polyurethanes to solid cover layers of all types wherein the improvement comprises treating the cover layers with an adhesive product comprising, based on the total weight,

(a) 1 to 10 percent by weight of an aminoalkyltrialkoxysilane,

(b) 1 to 20 percent by weight of a ...

PAGE 173

LEVEL 1 - 157 OF 225 PATENTS

4,337,216

<=2> GET 1st DRAWING SHEET OF 6

Jun. 29, 1982

Device in an evaporative cooler

INVENTOR: Korsell, Lars E. R., Stockholm, Sweden

 $\ldots$  [\*1] socket being telescopically inserted only into another single corresponding opening in an adjacent contact body.

[\*2] 2. In an evaporative cooler including, a casing, at least one contact body of the multi-layer type located in said casing and formed with channels existing between the layers and which all are passed by air, and means for supplying water to selected channels in said body from above the body, the improvement comprising ...

LEVEL 1 - 158 OF 225 PATENTS

174

4,305,670

Dec. 15, 1981

Liquid mixing device

INVENTOR: Moskowitz, Paul M., Brooklyn, New York Rushansky, Yuliy, Bronx, New York

 $\dots$  [\*1] surfaces of said disk extending in a plane perpendicular to the axis of rotation of said other shaft and being located symmetrically around said

said disk serving as a boundary layer type rotor in which boundary layer effects will occur along the surfaces of said disk during rotation of s disk, said boundary layer effect causing liquid to move in a downward, ... PAGE 175

LEVEL 1 - 159 OF 225 PATENTS

4,288,992

4 GET 1st DRAWING SHEET OF

Sep. 15, 1981

Curtain for open front freezer or refrigerator

INVENTOR: Eliason, Carlyle R., 905 W. Inkster, Kalamazoo, Michigan 49008

between said flexible sheet and strips when access to said access opening is not opening near at least one of said strip curtain and said air curtain means to the opposite wall of said cabinet to form an air layer-type thermal barrier flexible sheet curtain being extendible across said access ... [\*16] required.

17. The apparatus of claim 15, in which said upper front wall portion [\*17] 17. The app. of said cabinet is ...

LEVEL 1 - 160 OF 225 PATENTS

## 4 GET 1st DRAWING SHEET OF <=5>

Sep. 1, 1981

Textured surface polypropylene film

INVENTOR: Eustance, John W., So. Glens Falls, New York Hobbs, Stanley Y., Scotia, New York Carley, Emilie L., Hartford, New York

polypropylene film having one predetermined textured surface which is coextensively and uniformly covered by an overlapping pattern of fibroid irregularities comprising predominantly a stretched layer of Type I and Type II polypropylene crystal structure, and said film being characterized by a space factor of greater than about 5% and a haze measurement of greater than about

[\*2]  $\stackrel{|\uparrow\downarrow\downarrow|}{2}$ . The film of ... LEVEL 1 - 161 OF 225 PATENTS

4,265,386

7 GET 1st DRAWING SHEET OF

May 5, 1981

Torsional fluid damper system

INVENTOR: Levy, Avner, Irvine, California Karsh, Irving, Costa Mesa, California

... [\*12] a loop having a bight portion extending in contact with said circumferential wall of said container.

[\*13] 13. In a torsional fluid damper of the inertial mass and viscous fluid friction boundary layer type having a frequency response in the kHz region, the improvement comprising in combination:

means for containing an inertial mass and viscous fluid friction boundary layer comprising a rotatable container having an internal ...

LEVEL 1 - 162 OF 225 PATENTS

4,252,417

~ GET 1st DRAWING SHEET OF <=5>

Feb. 24, 1981

Liquid crystal display

177

INVENTOR: Scheffer, Terry J., Forch, Switzerland Zeller, Hans R., Birr, Switzerland

What is claimed as new and desired to be secured by Letters Patent of the United States is:

[\*1] 1. A liquid crystal display comprising:

two parallel plates having inside surfaces on which are formed layer-type electrodes and between which is disposed a liquid crystal mixture to form a liquid crystal cell, said plates having outside surfaces on which no polarizers are disposed;

said liquid crystal mixture comprising primarily ... LEVEL 1 - 163 OF 225 PATENTS

4,243,708

GET 1St DRAWING SHEET OF

Jan. 6, 1981

Metallized textured surface polypropylene film

INVENTOR: Eustance, John W., South Glens Falls, New York Hobbs, Stanley Y., Scotia, New York Carley, Emilie L., Hartford, New York

polypropylene film having one predetermined textured surface which is coextensively and uniformly covered by an overlapping pattern of fibroid irregularities comprising predominantly a stretched layer of Type I and Type II crystal structure, said film being characterized by a space factor of greater than about 5% and a haze measurement of greater than about 20%, and said film having an electrically ...

LEVEL 1 - 164 OF 225 PATENTS

4,231,754

Nov. 4, 1980

Chemiluminescent analytical device

INVENTOR: Vogelhut, Paul O., Mishawaka, Indiana

[\*4] a photoresponsive layer in contact with at least one surface of said second layer which is in contact with the first layer.

[\*5] 5. The test device of claim 4 wherein the photoresponsive layer is a photoresponsive imaging layer of the type which is permanently transformed by exposure to a light response in proportion to the amount of light emitted

4,229,095

GET 1st DRAWING SHEET OF 3

Oct. 21, 1980

Electro-optical color imaging apparatus

INVENTOR: Mir, Jose M., Webster, New York

... [\*5] produced so that the pixels of each successive strip are concurrently subjected to light of said different colors, sequentially and according to an image to be produced. [\*6] 6. Electro-optical color imaging apparatus for use with an imaging layer of the type which can record different light colors, said apparatus comprising: (a) means for disposing such an imaging layer in an imaging station of said apparatus:

(b), a plurality of discrete electro-optical ... LEVEL 1 - 166 OF 225 PATENTS

4,228,581

GET 1st DRAWING SHEET OF

Oct. 21, 1980

Method for producing semiconductor bodies having a defined edge profile which has been obtained by etching and is covered with a glass INVENTOR: Chadda, Madan M., Nu rnberg-Gaulnhofen, Federal Republic of Germany Maier, Reinhold, Nuremberg, Federal Republic of Germany

edge profile, said semiconductor bodies having a glass-covered defined edge profile, said semiconductor bodies being obtained by etching from a large-area semiconductor basic wafer having a sequence of layer-type zones of different conductivity type with at least one pn-junction and a surface oxide layer thereon, the steps comprising

applying a etch-resistant protective coating onto said surface oxide layer,

LEVEL 1 - 167 OF 225 PATENTS

183

4,223,234

<=2> GET 1st DRAWING SHEET OF 3

Sep. 16, 1980

Reduction of sparkle noise and mottling in CCD imagers

INVENTOR: Levine, Peter A., Trenton, New Jersey

... [\*4] B register responsive to applied multiple phase voltages for the storage in and transfer of charge along the channels of said B register;

said electrodes of said A and B registers being of the single layer type and comprising semiconductor material of one conductivity type and being separated from one another by "gaps" formed of semiconductor material of opposite conductfwity type; and

a control electrode insulated from the electrodes of the A register and PAGE 184

LEVEL 1 - 168 OF 225 PATENTS

4,206,372

<=2> GET 1st DRAWING SHEET OF 2

Jun. 3, 1980

Reduction of sparkle noise in CCD imagers

INVENTOR: Levine, Peter A., Trenton, New Jersey

applied multiple phase voltages for the storage in and transfer of charge along the channels of said B register, said electrodes being of the single layer type and comprising semiconductor material of one donductivity type and being separated from one another by semiconductor material

a CCD C register including a semiconductor formed with a ...

LEVEL 1 - 169 OF 225 PATENTS

4,160,684

<=2> GET 1st DRAWING SHEET OF 3

Jul. 10, 1979

Method of manufacturing a coalescing demister

INVENTOR: Berger, Jr., L. Joseph, Birmingham, Michigan Guequierre, Denis D., Birmingham, Michigan said cloth into place, thereby forming an anti-migration filter [\*#] ::: layer [\*5] 5. The method described in claim 4, and including the step of placing a screen type retainer inside said anti-migration layer of the type adapted to assert pressure on said anti-migration and said drain layers and keep them in intimate contact with one another and with said inner retainer.

[\*6] 6. The method described in claim 5, and including the ...

said cloth into place thereby forming an anti-migration filter ... [\*8] [\*9] 9. The method described in claim 8, and including the step of placing a screen-type retainer inside said anti-migration layer of the type adapted to assert pressure on said anti-migration and said drain layers and keep them in intimate contact with one another and with said inner coalescer retainer.

[\*10] 10. The method described in claim 9, and ... LEVEL 1 - 170 OF 225 PATENTS

4,150,186

<=2> GET 1st DRAWING SHEET OF 2

Apr. 17, 1979

Composite board structure and a method of and an apparatus for producing the board structure

INVENTOR: Kazama, Norio, Yokohama, Japan

... [\*7] copolymers, and ethylene-vinyl acetate copolymers.

[\*8] 8. A composite board structure as set forth in claim 1, in which the respective thermoplastic adhesive materials forming said first and second layers are of the types which are homogeneous to each other.

[\*9] 9. A composite board structure as set forth in claim 1, having three-dimensionally curved portion.

LÉVEL 1 - 171 OF 225 PATENTS

4,137,677

<=2> GET 1st DRAWING SHEET OF 1

Jan. 30, 1979

AGE 187

## Broadening the spatial frequency pass band of a thermoplastic layer

INVENTOR: Credelle, Thomas L., East Windsor, New Jersey Hannan, William J., Palm Beach Gardens, Florida Spong, Fred W., Lawrenceville, New Jersey

a οŧ multiplicity of frost frequencies, said undulations having a spatial frequency first and second surfaces respectively connected to a surface receive an interference pattern of light representative of an image, the improvement comprising a thermoplastic layer of the type that has a frost frequency inversely related to thickness, said thermoplastic layer having surface with undulations that cause said thermoplastic layer to have a substrate and to one surface of a photoconductor layer that is adapted to

... [\*4] less than 100 cycles per millimeter.

electrically conductive layer is connected to a surrow if that has a frost improvement comprising a photoplastic layer of the type that has a frost frequency inversely related to thickness, said photoplastic layer having a surface with undulations that cause said photoplastic layer to have a multiplicity of frost frequencies, said undulations having a spatial frequency PAGE [\*5] 5. In a holographic recording medium wherein one surface of an electrically conductive layer is connected to a surface of a substrate, the

LEVEL 1 - 172 OF 225 PATENTS

4,135,29

<=2> GET 1st DRAWING SHEET OF

 $\sim$ 

Jan. 23, 1979

Method for producing semiconductor devices with high reverse blocking capability

INVENTOR: Tursky, Werner, Schwabach, Eichvasen, Federal Republic of Germany Chadda, Madan, Nuremberg-Gaulnhofen, Federal Republic of Germany Schafer, Horst, Zirndorf, Federal Republic of Germany

Ø semiconductor disc of a first conductivity type with the devices having a high reverse blocking capability and having a sequence of at least three layer-type zones of different conductivity types, of which at least one is highly resistive, and at least one pn-junction, comprising the steps of:

forming grooves of a depth at least equal to the desired thickness of the highly resistive zone ... ... [\*1] said disc into sections of smaller areal expanse capable of being separated into individual semiconductor device containing wafers;

of at least three layer-type zones of different conductivity which form at least one pn-junction in each said section, and a zone of a single conductivity type which passes through the entire disc in the profile region of ... thereafter subjecting the disc to a diffusion process to produce a sequence

provide, in each said section, a sequence of three layer-type zones of alternating conductivity types with the zones adjacent both major surfaces being .. [\*2] a diffusion process includes diffusing an impurity which forms a zone of the opposite conductivity type into both major surfaces of said disc to of said<sup>#</sup>opposite conductivity type and an inner zone of said first conductivity type, and to provide a zone of said opposite ... LEVEL 1 - 173 OF 225 PATENTS

4,120,700

GET 1st DRAWING SHEET OF 2

Oct. 17, 1978

Method of producing p-n junction type elements by ionized cluster beam deposition and ion-implantation

INVENTOR: Morimoto, Kiyoshi, Mobara, Japan

produce an ohmic contact with the semiconductor layer of the one impurity type; substrate electrode being formed of a metal film such as can  $\cdots$  [\*1]

ionizing impurity atoms such as can form a semiconductor layer having the type of conductivity opposite to that of the semiconductor layer of the one impurity type;

accelerating the impurity ions by giving them kinetic energies;

implanting the impurity ions in the semiconductor layer of the one impurity type to form an ion- ... ... [\*2] substrate electrode being formed of a metal film such as can produce an ohmic contact with the semiconductor layer of the one impurity type;

ionizing impurity atoms such as can form a semiconductor layer having the type of conductivity opposite to that of the semiconductor layer of the one impurity type:

accelerating the impurity ions by giving them kinetic energies;

implanting the impurity ions in the semiconductor layer of the one impurity type to form an ion- ... ... [\*3] substrate electrode being formed of a metal film such as can produce an ohmic contact with the semiconductor layer of the one impurity type;

ionizing impurity atoms such as can form a semiconductor layer having the type of conductivity opposite to that of the semiconductor layer of the one impurity type;

accelerating the impurity ions by giving them kinetic energies;

implanting the impurity/ions in the semiconductor layer of the one impurity type to form an ion- ... ... [\*4] substrate electrode being formed of a metal film such as can produce an ohmic contact with the semiconductor layer of the one impurity type;

ionizing impurity atoms such as can form a semiconductor layer having the type of conductivity opposite to that of the semiconductor layer of the one impurity type;

PAGE

Pat. No. 4120700, \*4

accelerating the impurity ions by giving them kinetic energies;

implanting the impurity ions in the semiconductor layer of the one impurity type to form an ion- ...

LEVEL 1 - 174 OF 225 PATENTS

4,119,142

GET 1st DRAWING SHEET OF

Oct. 10, 1978

Arrangement for transferring heat from the exhaust air leaving an enclosed volume to the input air supplied to said volume

INVENTOR: Margen, Peter Heinrich Erwin, Nykoping, Sweden

in said circuit, arranged in said supply conduit, a second heat exchanger in said circuit, arranged in said exhaust conduit, the improvement comprising a heat store of the stratified liquid-layer type, the hot side of which is coupled to the hot side of the circuit and the cold side of which is coupled to the cold side of said circuit, valve means for deflecting part of the hot circuit liquid into said store during normal ...

... [42] least one by-pass line arranged in said circuit in parallel with said second heat exchanger;

means for controlling the liquid flow through said second heat exchanger;

layer type heat store means for supplying heat to said first heat exchanger in the form of heat extracted from said exhaust air, while said second heat exchanger is disconnected for defrosting, having the ...

191

4,096,389

<=2> GET 1st DRAWING SHEET OF 5

Jun. 20, 1978

Apparatus for minimizing radiation exposure and improving resolution in radiation imaging devices

INVENTOR: Ashe, John B., Palatine, Illinois Williams, Gwilym H., Palatine, Illinois Sypal, Kenneth L., Glen Ellyn, Illinois

... [\*6] improvement as defined in claim 1, wherein said sandwich assembly comprises a plurality of substantially equally spaced concentric cylindrical layers of alternating attenuation and spacing material, each said respective layer type being substantially uniform in length and in thickness, whereby a circular beam pattern is produced.

7. The improvement as defined in claim 1, wherein said sandwich assembly comprises a layer of

LEVEL 1 - 176 OF 225 PATENTS

4,092,663

<=2> GET 1st DRAWING SHEET OF 2

. . . . . May 30, 1978

Semiconductor device

INVENTOR: Schafer, Horst, Zirndorf-Nuremberg, Germany, Federal Republic of

I claim:

comprising a semiconductor wafer having at least two layer type zones of alternatingly opposite conductivity type and different doping concentrations and forming a pn junction therebetween which intersects the edge surface of the semiconductor wafer, the higher doped of said at least two zones forming ... 1. In a semiconductor device with a high blocking capability [\*1]

LEVEL 1 - 177 OF 225 PATENTS

4,087,159

<=2> GET 1st DRAWING SHEET OF 15

May 2, 1978

INVENTOR: Ulrich, Reinhard, Leonberg-Silberberg, Germany, Federal Republic of

... [\*24] provided at an input window (E1) in FIG. 31) taking up the one half of the input surface of the waveguide.

[\*25] 25. An imaging system according to claim 24, characterized by the feature that the waveguide is a layer type waveguide, which has a different thickness in the region (PS) of the input window (E1) than in the other regions.

[\*26] 26. An imaging system according to claim 24, characterized by the feature that the phase shift device ( ... LEVEL 1 - 178 OF 225 PATENTS

4,084,863

GET 1st DRAWING SHEET OF 1

Apr. 18, 1978

Bearing and bearing liner having a compliant layer

INVENTOR: Capelli, Alfred J., Palos Verdes Peninsula, California

... [\*1] height of the projection on the substrate.

[\*2] 2. In a bearing including a substrate having an irregular surface and further including a thin and deformable wear resistant layer having a wear surface wherein the wear resistant layer is of the type which would be deformed by the irregular surface of the substrate upon loading of the wear resistant layer against the irregular surface, the wear resistant layer including a porous backing member and particles of a lubricant ...

~ GET 1st DRAWING SHEET OF

Jul. 19, 1977

Avalanche photodiode

INVENTOR: de Cremoux, Baudouin, Paris, France

 $\dots$  [\*4] 3, wherein the thickness of the third layer is sufficiently thin to be transparent to the radiation, the vadiation being absorbed in the first

[\*5] 5. A diode as claimed in claim 4, wherein said first layer has type n-conductivity, said second and third layers having type p-conductivity, the first layer having a doping concentration of the order of 10<16 at/cm<3>,

said second and said third layers having doping concentrations of the order of 10<19 > at/cm<3> , the thickness of the ...

... [\*6] 3, wherein said third layer is sufficiently thick to absorb the radiation to be detected.

[\*7] 7. A diode as claimed in claim 6, wherein the first layer has p-type conductivity, the second and third layers have type n-conductivity, the doping concentrations being of the order of 10<18 > at/cm<3 > and 10<16 > at/cm<3>, respectively. 8. A diode as claimed in claim 1, wherein the layers are made of PAGE 197

LEVEL 1 - 180 OF 225 PATENTS

4,019,843

<=2> GET 1st DRAWING SHEET OF 4

Apr. 26, 1977

Film blowhead for producing tubular film

INVENTOR: Zimmermann, Werner Josef, Lengerich of Westphalia, Germany

... [\*3] in each of said pairs disposed in a common radial plane and with the radial planes in parallel relationship.

[\*4] 4. A film blowhead according to claim 1 wherein the film blowhead is of multi-layer type and wherein said air inlet and outlet tubes are disposed between said axial passage portions of said distributing passages in at least one radial plane.

LEVEL 1 - 181 OF 225 PATENTS

4,015,034

<=2> GET 1st DRAWING SHEET OF 2

Mar. 29, 1977

Register for index marking article

INVENTOR: Smolen, Benjamin Edward, 1501 Broadway, New York 10036

... [\*2] said carrier includes an adhesive surface on the face opposite said release surface for securing said carrier to a said sheet. [\*3] 3. Article in accordance with claim 1 wherein said adhesive layer is of the type having an initial low tack which is rendered highly adherent responsive to localized high pressure such as exerted by a stylus scanned in registry therewith.

AGE 198

[\*4] 4. Article in accordance with claim 3 wherein said ... E 199

LEVEL 1 - 182 OF 225 PATENTS

4,012,817

<=2> GET 1st DRAWING SHEET OF 1

Mar. 22, 1977

Method of making a capacitor

INVENTOR: Preissinger, Karl-Heinz, Taufkirchen, Germany, Federal Republic of Wehnelt, Ulrich, Starnberg, Germany, Federal Republic of

We claim as our invention:

[\*1] 1. A method for producing a layer type capacitor comprising the steps of

coating an adhesion-imparting layer in a dissolved state onto a first covering foil,

applying a first conductive layer with pores therein into the exposed surface of said adhesion- ...

 $\ldots$  [\*1] sides by pressing heated leads at least at one point through said

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covering foil and the respective of said layers into said first covering foil.

[\*2] 2. A method for producing a layer type capacitor in accordance with claim 1 wherein the hardening glue is added to the adhesion-imparting layer in a concentration which does not prevent activation of said adhesion-imparting layer by penetration of the dielectric solvent ...

LEVEL 1 - 183 OF 225 PATENTS

3,969,232

GET 1st DRAWING SHEET OF 1 <=2>

Jul. 13, 1976

Bearing and bearing liner wear resistant compliant layer

INVENTOR: Turner, Peter H., Burbank, California

mixture including the adhesive. ... [\*2] [\*3] 3. In a bearing including a substrate having an irregular surface,

layer against the irregular surface, the first wear resistant layer including a first thin wear resistant layer having a first wear surface wherein the first wear resistant layer is of the type which would be deformed by the irregular surface of the substrate upon loading of the first wear resistant first porous backing member and ... LEVEL 1 - 184 OF 225 PATENTS

GET 1st DRAWING SHEET OF

Sealing means for wind instruments

INVENTOR: Nelson, Robert E., Los Angeles, California Gilbert, Robert D., Los Angeles, California

... [\*4] layer.

[\*5] 5. In the sealing means of claim 4 wherein said first layer is Volara material and said second layer is Volite material.

[\*6] 6. In the sealing means of claim 5 wherein said first layer is Type Volāra material. [\*7] 7. In the sealing means of claim 6 wherein a third layer of material having indicia thereon is adhered to said second layer, said third layer being fixedly secured to said ...

... [\*12] first layer is laminated to said second layer.

[\*13] 13. The pad of claim 12 wherein said first layer is Volara material and the second layer is Volite material.

14. The pad of claim 13 wherein said first layer is Type A Volara [\*14] material.

[\*15] 15. The pad of claim 14 wherein a third layer of material having indicia thereon is adhered to said second layer. LEVEL 1 - 185 OF 225 PATENTS

3,956,624

0 GET 1st DRAWING SHEET OF

May 11, 1976

Method and device for the storage and multiplication of analog signals

INVENTOR: Audaire, Luc, St-Nizier-du-Moucherotte, France Borel, Joseph, Echirolles, France LE Goascoz, Vincent, Claix, France Poujois, Robert, Grenoble, France

a multiplication of two corresponding terms is performed by recording the signal which is proportional to one sample of said function in a memory of the multiple dielectric layer type and by applying a given voltage to the gate of said transistor so as to generate a signal which is a linear function of the threshold voltage which is in turn a linear function of the ordered series is derived from the sampling of a function, wherein

LEVEL 1 - 186 OF 225 PATENTS

3,949,463

<=2> GET 1st DRAWING SHEET OF 2

Apr. 13, 1976

Method of applying an anti-reflective coating to a solar cell

INVENTOR: Lindmayer, Joseph, Bethesda, Maryland Allison, James F., Silver Springs, Maryland

.. [\*1] as a solar cell, an anti-reflective coating and a desired pattern of a metal electrode for current collection, comprising the steps of:

a, coating said surface with a first metal layer of the type which can be oxidized to form said anti-reflective coating,

b. forming on top of said coating a metal electrode having said desired pattern; said first metal layer having parts thereof exposed which are ... PAGE 204

LEVEL 1 - 187 OF 225 PATENTS

1,141

3,939,642

<=2> GET 1st DRAWING SHEET OF 4

Feb. 24, 1976

Electronic timepiece semiconductor intergrated circuit

INVENTOR: Morozumi, Shinji, Nagano, Japan

... [\*4] substrate is formed of a material selected from the group consisting of sapphire, spinel, silicon oxide or titanium oxide.

[\*5] 5. An electronic timepiece as claimed in claim 3 wherein the P-channel transistors are depletion-layer type transistors and the N-channel transistors are reverse-layer type transistors.

[\*6] 6. An electronic timepiece as claimed in claim 3, wherein said P-channel and N-channel transistors are reverse-layer channel transistors.

7. An electronic timepiece as claimed in ... LEVEL 1 - 188 OF 225 PATENTS [\*7]

3,936,730

<=2> GET 1st DRAWING SHEET OF 1

205

Feb. 3, 1976

Insulation test apparatus including improved means for simultaneous display

INVENTOR: Pittman, Paul F., Pittsburgh, Pennsylvania

....[\*3] branches is a current transformer and said means for displaying signals is an oscilloscope.

4. The subject matter of claim 2 wherein:

said switching means comprises a plurality of semiconductor switching devices of the four layer type connected in a series string and provided with means to trigger said series string to conduction to effect discharge of said capacitor.

5. The subject matter of claim 1 wherein: [\*5] said means for displaying ... LEVEL 1 - 189 OF 225 PATENTS

3,930,903

GET 1st DRAWING SHEET OF <=5

Jan. 6, 1976

Stabilized superconductive wires

INVENTOR: Randall, Robert N., Wayland, Massachusetts
Wong, James, Wayland, Massachusetts

What is claimed is:

[\*1] 1. Superconductive multi-filament wire product comprising,

means defining a copper matrix with a plurality of spaced filaments therein,

each of the filaments comprising a layer therein of type II superconducting intermetallic compound of Beta-Wolfram structure, being the diffusion reaction product of source filaments comprising a cross-section multilayer configuration of a refractory metal layer each containing ...

3,929,849

GET 1St DRAWING SHEET OF

Dec. 30, 1975

Tetraalkyl phosphonium aluminosilicates

. .

INVENTOR: Oswald, Alexis A., Mountainside, New Jersey

What is claimed is:

- 1. Tetra-alkyl phosphonium clays of layer and chain type structure. [4]
- 2. Tetra-alkyl phosphonium clays of layer type structure [\*2]
- [\*3] 3. The compositions of claim 2 wherein said layer type clay is a montmorillonite.
- [\*4] 4. Tetra-alkyl phosphonium clays of the formula [R4P + ] Clay

wherein R is a C1 to C100 aliphatic hydrocarbyl group, and the clay is a negatively charged aluminosilicate of layer and chain ...

... [\*8] C1 to C7 low aliphatic groups and C8 to C100 high aliphatic groups in such a manner that if R¹ is low, R" should be high and the reverse; Clay - is a negatively charged layered aluminosilicate of layer type structure.

[\*9] 9. The composition of claim 8 wherein the high aliphatic groups equal C8 to C40 alkyl and the low C1 to C7 aliphatic groups are selected from the group consisting of alkyl, alkenyl and alkinyl.

[\*10] 10. The composition of ... LEVEL 1 - 191 OF 225 PATENTS

208

PAGE

3,922,777

GET 1st DRAWING SHEET OF

Dec. 2, 1975

Process for the production of layer circuits with conductive layers on both sides of a ceramic substrate

INVENTOR: Weitze, Artur, Pullach, Germany, Federal Republic of Leskovar, Peter, Munich, Germany, Federal Republic of

We claim as our invention:

[\*1] 1. A process for the production of layer-type printed circuits having conductive layers on both sides of a ceramic substrate which comprises providing an aperture in a green ceramic substrate, inserting into said aperture, a high melting metal pin having ...

LEVEL 1 - 192 OF 225 PATENTS

<=2> GET 1st DRAWING SHEET OF 5

209

Integrated IGFET bucket-brigade circuit

INVENTOR: Adam, Fritz G., Freiburg, Germany, Federal Republic of Obermeier, Cornelius, Freiburg, Germany, Federal Republic of Scheffer, Gerhard, Denzlingen, Germany, Federal Republic of Wilmsmeyer, Klaus, Denzlingen, Germany, Federal Republic of

... [\*1] sources of clock pulses, said first source coupled to said even-numbered transistors and said second source coupled to said odd-numbered transistors; and a row of field-effect transistors of the depletion-layer type having source and drain regions, said field-effect transistors employing gate electrodes on an insulated-gate layer, wherein the last transistor in said row is provided with an electrical terminal to which said source of operating voltage is ...

LEVEL 1 - 193 OF 225 PATENTS

3,910,862

GET 1st DRAWING SHEET OF

Oct. 7, 1975

Stabilized superconductors

INVENTOR: Wong, James, Wayland, Massachusetts

What is claimed is:

 Superconductive multi-filament wire product comprising, [<del>\*</del>] means defining a bronze matrix with a plurality of spaced filaments therein,

each of the filaments comprising a layer of type II superconducting intermetallic compound of Beta-Wolfram structure being the diffusion reaction product of a first elemental component derived from said bronze matrix and of second elemental component derived from source ... LEVEL 1 - 194 OF 225 PATENTS

3,895,336

GET 1st DRAWING SHEET OF

Jul. 15, 1975

Transformer core with composite offset V-miter and step ioint

INVENTOR: Pitman, Frank A., Rome, Georgia

211

I claim:

[\*1] 1. In a transformer core of the stacked flat-layer type having a plurality of flat, laminated, layered members of equal width, each of said members including: two generally rectangular shaped, parallel, spaced-apart, outside leg members beveled at each end;

LEVEL 1 - 195 OF 225 PATENTS

3,895,335

GET 1st DRAWING SHEET OF 1

Jul. 15, 1975

Series/parallel connected single phase power transformer

INVENTOR: Manimalethu, Abraham I., Peru, Massachusetts

consists of two high voltage windings and two low voltage ... [\*2] windings.

- [\*3] 3. A single phase electrical power transformer as defined in claim 2 wherein said high voltage windings are of the layer type, the low voltage windings between high voltage windings is of the helical type and the remaining low voltage winding is of the layer type.
- [\*4] 4. A single phase electrical power transformer as defined in claim 2 wherein said high voltage windings are of the layer type, the low voltage windings between high voltage windings is of the helical type and the remaining low voltage winding is of the disc type.
- [\*5] 5. A single phase electrical power transformer as defined in claim 2 wherein said high voltage windings are of the disc type, the low voltage winding between high voltage windings of the helical type and the remaining low voltage winding is of the layer type.
- [\*6] 6. A single phase electrical power transformer as defined in claim 2 wherein said high voltage windings are of the disc type, the low voltage winding between the high voltage windings is of the ... LEVEL 1 - 196 OF 225 PATENTS

3,892,655

Jul. 1, 1975

Layered clay minerals, catalysts, and processes for using

INVENTOR: Hickson, Donald A., Richmond, California

What is claimed is:

[\*1] 1. A hydroconversion process comprising contacting a hydrocarbon feedstock at conventional hydroconversion conditions with a catalyst comprising: (1) a layer-type trioctrahedral, clay-like mineral, and (2) at least one hydrogenation component, said mineral having prior to dehydrating and calcining of said catalyst, the empirical formula:
MgO: sSiOZ: aAl2O3: bAB: xH2O

wherein the layer-lattice ...

claim 1 wherein said hydrogenation component comprises platinum. [<del>\*</del>]

catalyst comprising a layer-type trioctahedral, clay-like mineral, said mineral having prior to dehydrating and calcining of said catalyst the empirical [\*10] 10. A catalytic conversion process comprising contacting a hydrocarbon feedstock at conventional catalytic conversion conditions with a

MgO : SS102 : aA1203 : bAB : xH20

wherein the layer-lattice structure is composed of said silica, said ... DAGF 214

LEVEL 1 - 197 OF 225 PATENTS

3,888,678

Jun. 10, 1975

Method for adjusting triboelectric charging characteristics of materials

INVENTOR: Bailey, Jr., William J., Rochester, New York Houle, James F., Rochester, New York Van Norman, Gilden R., Rochester, New York ... [\*50] agent has the following empirical structure:

[\*51] 51. The film base element of claim 50 further including Saponin.

[\*52] 52. A film base element suitable for the reception of at least one radiation sensitive layer and of the type subject to triboelectric charging upon impact and dissociation with another usually dissimilar material said element having a surface thereof modified against generation of triboelectrical charges sufficient in electrical potential to cause static ...
LEVEL 1 - 198 OF 225 PATENTS

,887,454

<=2> GET 1st DRAWING SHEET OF 1

Jun. 3, 1975

Layered clay minerals and processes for using

INVENTOR: Hickson, Donald A., Richmond, California

What is claimed is:

feedstock at conventional hydroconversion conditions with a catalyst comprising:
(1) a layer-type dioctahedral, clay-like mineral, and (2) at least one hydrogenation component, said mineral having prior to dehydrating and calcining of said catalyst, the empirical formula:
Mg0 : sSi02 : aAl203 : bAB : xH20 1. A hydroconversion process comprising contacting a hydrocarbon

wherein the layer-lattice ...

claim 1 wherein said hydrogenation component comprises platinum. [6\*] ...

[\*10] 10. A catalytic conversion process comprising contacting a hydrocarbon feedstock at conventional catalytic conversion conditions with a catalyst comprising a layer-type dioctahedral, clay-like mineral, said mineral having prior to dehydrating and calcining of said catalyst the empirical formula:

MgO : SS102 : aA1203 : bAB : xH20

wherein the layer-lattice structure is composed of said silica, said ...

LEVEL 1 - 199 OF 225 PATENTS

3,884,539

<=2> GET 1st DRAWING SHEET OF 1

May 20, 1975

Method of making a multialkali electron emissive layer

INVENTOR: Sommer, Alfred Hermann, Princeton, New Jersey

I claim:

[\*1] 1. A method of activating a multialkali electron-emissive cathode layer, of the type wherein a layer of antimony is exposed at elevated temperature, within an evacuated body, to vapors of a plurality of alkali metals including cesium, to form an electron-emissive compound, wherein the improvement comprises:

exposing the cathode layer to ... LEVEL 1 - 200 OF 225 PATENTS

PAGE 217

Production of synthetic silicate minerals

INVENTOR: Hoffman, George W., Houston, Texas Blankenship, H. Michael, Houston, Texas Granquist, William T., Houston, Texas

Having described the invention, we claim:

[\*1] 1. The process of producing a 2:1 layer-type clay-like mineral product having the empirical formula: nSi02:Al203:mAB:xH20

where the layer lattices comprise said silica, said alumina, and said B, and where

n is from 1.7 to 3.0,

m is from 0.2 to 0.6,

PAGE 218

LEVEL 1 - 201 OF 225 PATENTS

3,864,931

<=2> GET 1st DRAWING SHEET OF 1

Feb. 11, 1975

PROCESS AND APPARATUS FOR FOOD FREEZING

INVENTOR: Guttinger, Manfred, Leinfelden, Germany, Federal Republic of

... [\*11] second flow than for said first flow.

12. Process for freezing foodstuffs comprising the successive steps [\*12] of:

extremely difficult for the medium to flow through the layer and through the horizontal support, which layer is of the type which would exert such a resistance against the through flow of a medium from above that it would be support which has a plurality of spaced openings extending therethrough placing foodstuffs in a substantially flat layer on a substantially

PAGE 219

LEVEL 1 - 202 OF 225 PATENTS

3,864,726

<=2> GET 1st DRAWING SHEET OF 1

北京

Feb. 4, 1975

## CONTROLLABLE SEMICONDUCTOR RECTIFIER

INVENTOR: Semikron Gesellschaft fur Gleichrichterbau und Elektronid m.b.H., Zirndorf, Germany, Federal Republic of

I claim:

which is adjacent to the one of the outer zones of said semiconductor body which monocrystalline semiconductor body having planar major outer surfaces and four layer-type zones of alternatingly opposite conductivity types with the one of the inner zones of said semiconductor body which serves as the base zone, and 1. In a controllable semiconductor rectifier device including: a serves as the emitter ...

LEVEL 1 - 203 OF 225 PATENTS

3,858,236

GET 1st DRAWING SHEET OF 2

Dec. 31, 1974

FOUR LAYER CONTROLLABLE SEMICONDUCTOR RECTIFIER WITH IMPROVED FIRING PROPAGATION SPEED

INVENTOR: Schafer, Horst, Zirndorf, Germany, Federal Republic of Herbing, Lothar, Nurnberg, Germany, Federal Republic of

We claim:

monocrystalline semiconductor body having four layer-type zones of alternatingly opposite conductivity types and with the one of the inner zones of said semiconductor body which borders on the one of the outer zones of said semiconductor body which serves as the emitter zone of the device having a 1. In a controllable semiconductor rectifier device including: a portion whereof which is to ...

LEVEL 1 - 204 OF 225 PATENTS

3,854,983

Dec. 17, 1974

FLAMEPROOF COVERING MATERIAL, SUCH AS TICKING

INVENTOR: Brodnyan, John G., Langhorne, Pennsylvania

I claim:

[\*1] 1. A cover fabric of composite-layer type comprising a light-weight woven or damask fabric, a soft, flexible layer adhered thereto formed of a

222 crushed foam of a polymeric material and a metal-containing coating of about 0.5

LEVEL 1 - 205 OF 225 PATENTS

3,849,217

<=2> GET 1st DRAWING SHEET OF 6

Nov. 19, 1974

METHOD OF MANUFACTURING HIGH FREQUENCY DIODE

INVENTOR: Kroger, Harry, Sudbury, Massachusetts Potter, Curtis N., Holliston, Massachusetts

We claim:

[\*1] 1. The method of making a high frequency diode device from a body of semiconductor material having type n + conductivity and having an epitaxial layer of type n conductivity, the method comprising:

forming a layer of type p conductivity material at a surface of said epitaxial layer,

forming a metal layer of chromium over said surface,

forming a metal layer of gold over said chromium layer,

... [\*4] contiguous metal ring layers by etching, and

removing said mask.

[\*5] 5. The method of making a high frequency diode device from a body of semiconductor material having type n + conductivity and an epitaxial layer having type n conductivity, the method comprising:

forming a layer of type p conductivity material at a first free surface of said epitaxial layer,

forming a base layer of gold at a second free surface of said type n + semiconductor material,

forming ...

LEVEL 1 - 206 OF 225 PATENTS

3,844,979

0ct. 29, 1974

LAYERED CLAY MINERALS, CATALYSTS, AND PROCESSES FOR USING

PAGE 223

INVENTOR: Hickson, Donald A., Richmond, California

What is claimed is:

[\*1] 1. A layer-type, trioctahedral, clay-like mineral having the empirical
formula
Mg0 : sSi02 : aAl203 : bAB : xH20

wherein the layer-lattice structure is composed of said silica, said alumina, said magnesia, said A and B, and wherein

s is from ...

... [\*5] hydrogen form, wherein s = 1.166 a = 0.08, and said mineral having after calcination a fluoride content of from 1 to 3 weight percent.

[\*6] 6. A catalytic cracking catalyst comprising dehydrated, calcined, layer-type, trioctahedral, clay-like mineral having prior to dehydration and calcining of said catalyst the empirical formula MgO: sSiO2: aAl2O3: bAB: xH2O

wherein the layer-lattice structure is composed of said silica, said alumina said ...

 $\dots$  [\*7] dehydrated mineral is composited with an amorphous inorganic oxide.

 $[*8] \ 8.$  The catalytic cracking catalyst of claim 6 wherein said dehydrated mineral is composited with a zeolite.

9. A catalyst composite comprising: [43] A. a layer-type, trioctahedral, clay-like mineral having prior to dehydration and calcining of said catalyst the empirical formula MgO : sSiO2 : aAl203 : bAB : xH20

wherein the layer-lattice structure is composed of said silica, said alumina

LEVEL 1 - 207 OF 225 PATENTS

3,844,978

GET 1st DRAWING SHEET OF

Oct. 29, 1974

LAYERED CLAY MINERALS AND PROCESSES FOR USING

INVENTOR: Hickson, Donald A., Richmond, California

What is claimed is:

1. A layer-type, dioctahedral, clay-like mineral having the empirical MgO : sSi02 : aA1203 : bAB : xH20 [\*1] formula

wherein the layer-lattice structure is composed of said silica, said alumina, said magnesia, said A and said B, and wherein

:

... [\*5] hydrogen form, wherein s=3.28, a=0.74, and said mineral having after calcination a fluoride content of from 1 to 3 weight percent.

[\*6] 6. A catalytic cracking catalyst comprising dehydrated, calcined, layer-type, dioctahedral, clay-like mineral having prior to dehydration and calcining of said catalyst the empirical formula
MgO: sSiO2: aAl203: bAB: xH20

wherein the layer-lattice structure is composed of said silica, said alumina,

dehydrated mineral is composited with an amorphous inorganic ••• [\*7] oxide. [\*8] 8. The catalytic cracking catalyst of claim 6 wherein said dehydrated mineral is composited with a zeolite.

[\*9] 9. A catalyst composite comprising:

A. a layer-type, dioctahedral, clay-like mineral having prior to dehydration and calcining of said catalyst the empirical formula MgO : sSiO2 : aAl2O3 : bAB : xH2O

wherein the layer-lattice structure is composed of said silica, said alumina, PAGE

LEVEL 1 - 208 OF 225 PATENTS

3,818,248

<=2> GET 1st DRAWING SHEET OF 2

Jun. 18, 1974

SERIALLY CONNECTED SEMICONDUCTOR SWITCHING DEVICES
SELECTIVELY CONNECTED FOR PREDETERMINED VOLTAGE BLOCKING AND
RAPID SWITCHING

INVENTOR: Pittman, Paul F., Pittsburgh, Pennsylvania

 $\dots$  [\*1] minimizes the turn on time of said first number of devices.

[\*2] 2. The subject matter of claim 1 wherein: said voltage varies over a range of at least an order of magnitude; said switching devices are of the four layer type; said means for selectively connecting is such that said second number of said swtiching devices is in two groups of approximately equal size at the ends of the series connection.

\*3] 3. The subject matter of claim 1 wherein: ... LEVEL 1 - 209 OF 225 PATENTS

3,816,343

Jun. 11, 1974

KAOLINITE COATED WITH SYNTHESIZED LAYER-TYPE SILICATE MINERALS

INVENTOR: Hoffman, George W., Houston, Texas Granquist, William T., Houston, Texas

laving described the invention, we claim:

[\*1] 1. The process of preparing a synthetic layer-type mineral-kaolinite complex which consists in commingling kaolinite with a reaction mixture consisting essentially of:

water

a minor proportion of alumina;

silica in the molar ratio to said alumina of 2.7 to 3.3; and

•

... [\*1] alumina of 0.2 to 0.6;

thereafter autoclaving the mixture thus formed at a temperature within the range of 2800 to 315oC. for a period of time sufficient for said reaction mixture to be converted to a layer-type clay-like mineral;

and cooling said mixture and recovering said complex therefrom.

- [\*2] 2. The process in accordance with claim I wherein the weight ratio of solids in said reaction mixture to said kaolinite is within the range of from 5 : 1 to 1 : 5.
- [\*3] 3. A complex consisting essentially of particles of kaolinite coated with a layer-type mineral having the empirical formula: nSi02:A1203:mAB:xH20

where the layer lattices comprise said silica, said alumina, and said B, and

- n is from 2.4 to 3.0,
- m is from 0.2 to 0.6,
- A is one equivalent of an ...
- ... [\*3] density than a mechanical mixture of the same said components of said clay-like mineral in said kaolinite.
- 227 [\*4] 4. A complex in accordance with claim 3 in which the weight ratio of said layer-type mineral to said kaolinite is within the range of from 5:1 to 1:5.

LEVEL 1 - 210 OF 225 PATENTS

3,761,171

<=2> GET 1st DRAWING SHEET OF 3

Sep. 25, 1973

NEGATIVE-POSITIVE, POSITIVE-POSITIVE EXPOSURE STATION

INVENTOR: Fields, Gary D., Parker, Colorado

... [\*15] comprising:

means for supporting the photosensitive surface for exposure;

- a layered sandwich structure which includes in order:
- a first transparent electrode;
- a photoconductive layer;
- a liquid crystal layer of the type having the capacity to store an image at least temporarily; and
- a second transparent electrode;
- means for applying a first potential between said electrodes during formation of a temporary image in said ...
- means for supporting the charged photoconductive surface for ... [\*16] exposure;
- a layered sandwich structure which includes in order:
- a first transparent electrode;
- a photoconductive layer;

a liquid crystal layer of the type having the capacity to store an image at least temporarily; and

a second transparent electrode;

means for applying a first potential between said electrodes during formation a temporary image in said ... LEVEL 1 - 211 OF 225 PATENTS of

3,720,847

GET 1st DRAWING SHEET OF 3

Mar. 13, 1973

POWER CURRENT CRYOTRON WITH FLAT GATE CONDUCTOR

INVENTOR: Massar, Ernst, Erlangen, Germany, Federal Republic of

I claim:

[\*1] 1. A power current cryotron comprising an insulating member and a layer type gate conductor superconducting layer on the insulating member, said layer having a thickness in the order of magnitude of the depth of penetration of a magnetic field into the superconducting layer, said insulating member and said layer ... ... [\*1] during operation of said cryotron adjacent portions of said layer conduct current in opposite directions.

[\*2] 2. A power current cryotron comprising a tubular insulating member having an axis and a layer type gate conductor superconducting layer on said insulating member, said layer having a thickness in the order of magnitude of the depth of penetration of a magnetic field into the superconducting layer, said insulating member and said ... LEVEL 1 - 212 OF 225 PATENTS

3,719,535

GET 1st DRAWING SHEET OF

Mar. 6, 1973

HYPERFINE GEOMETRY DEVICES AND METHOD FOR THEIR FABRICATION

INVENTOR: Zoroglu, Demir S., 4917 North 73rd Street, Apt. 9, Scottsdale, Arizona 85251

... [\*1] sequence of steps and the use of materials for minimizing the number of steps required, comprising the steps of:

providing a semiconductor body of a first type of conductivity and having an

forming a first passivating layer of the type operating to act as a diffusion barrier on said upper surface;

upper surface:

forming a plurality of apertures in said passivating layer which are aligned each to the other:

forming a second passivating layer of the type through which conductivity type determining impurities pass over said first layer and said exposed surface of said semiconductor body;

forming a third passivating layer of the type operating to act as a diffusion barrier over said second layer;

patterning said third layer such as to form at least one aperture overlying a selected aperture in said ...

... [\*6] steps and through the use of materials for minimizing the number of steps required, comprising the steps of:

providing a semiconductor body of a first type of conductivity and having upper surface: forming a first passivating layer of the type operating to act as a diffusion barrier on said upper surface;

forming a plurality of apertures in said passivating layer which are aligned each to the other for exposing an equal plurality of surface ... E 230

LEVEL 1 - 213 OF 225 PATENTS

3,716,969

0 GET 1st DRAWING SHEET OF

Feb. 20, 1973

CONTINUOUS MOVING LAYER TYPE ADSORPTION DEVICE

INVENTOR: Maeda, Isamu, Niihama-shi, Japan

What I claim is:

[\*1] 1. A continuous moving layer type adsorption device employed in a gas desulfurization system, comprising:

a. an adsorption vessel main body filled with activated charcoal, and

b. a rectifying device, said adsorption vessel main ... LEVEL 1 - 214 OF 225 PATENTS

PAG

231

Jan. 30, 1973

# MULTI-LAYER COLOR PHOTOGRAPHIC SILVER HALIDE LIGHT-SENSITIVE MALERIALS

NVENTOR: Hayashi, Jun, Kanagawa, Japan

Sato, Akira, Kanagawa, Japan

What is claimed is:

[\*1] 1. A multi-layer type color photographic light-sensitive material characterized in that a merocyanine dye having the following general formula [I]

wherein X is a member selected from the group consisting of a sulfur atom, a selenium ...

benzselenazoles naphthoselenazoles, benzimidazoles, naphthoimidazoles, 2-quinolines, 2-pyridines, and indolenines, is incorporated in at least one layer of said multi-layer type color photographic light-sensitive material, said multi-layer type color photographic light-sensitive material, in ... [\*1] consisting of thiazolines, thiazoles, benzthiazoles, naphthothiazoles, oxazoles, benzoxazoles, naphthoxazoles,

1. a support,

[\*2] 2. a subbing layer,

[\*3] 3. a red sensitive silver halide emulsion layer,

[\*4] 4.a..

... [\*5] I to 10.0 mole percent, the remaining silver halide emulsion layers containing a silver halide selected from the group consisting of AgBr, AgI, AgCIBr, AgCII, AgIBr and AgCIIBr.

[\*2] 2. The multi-layer type color photographic light-sensitive material as claimed in claim 1 wherein each of R1, R2 and R3 is selected from a group consisting of a hydrogen atom, a methyl group, an ethyl  $\dots$ 

... [\*2] group, a sulfobutyl group, a 4-carboxyphenethyl group, a 4-sulfophenethyl group, a phenyl group, a 4-carboxyphenyl group, and a 4-sulfophenyl group.

Pat. No. 3713828, \*2

[\*3] 3. The multi-layer type color photographic light-sensitive material as claimed in claim 1 wherein the heterocyclic ring completed by Z is selected from the group consisting of the thiazolines, thiazoles, benzthiazoles, naphthothiazoles, oxazoles, benzoxazoles, naphthothiazoles, selenazoles,

'AGE 23;

benzselenazoles, naphthoselenazoles, indolenines, benzimidazoles, naphthoimidazoles, 2-quinolines and 2-pyridines.

- [\*4] 4. The multi-layer type color photographic light-sensitive material as claimed in claim 1, wherein said merocyanine dye is incorporated in at least one of the layers consisting of the silver halide emulsion layers and the layers adjacent to the silver halide emulsion layers.
- [\*5] 5. The multi-layer type color photographic light-sensitive material as claimed in claim 1, wherein, after development, cyan, magenta, and yellow images are formed in the red-sensitive, the green-sensitive, and the blue-sensitive silver halide emulsion layers, respectively.
- [\*6] 6. The multi-layer type color photographic light-sensitive material as claimed in claim 1, wherein the silver halide is selected from the group consisting of silver bromide, silver iodide, silver chloride, silver chlorobromide, silver iodobromide, and silver chloro-iodobromide.
- 7. The multi-layer type color photographic light-sensitive material as claimed in claim 1, wherein said merocyanine dye has the formula
- 8. The multi-layer color photographic light-sensitive material claimed in ...

LEVEL 1 - 215 OF 225 PATENTS

PAGE

FRONT CONTACTED ELECTRICAL COMPONENT

Jan. 9, 1973

GET 1st DRAWING SHEET OF

INVENTOR: Behn, Reinhard, Balanstr. 95, Munich, Germany, Federal Republic of Gottlob, Heinrich, Annahofstr. 25, Regensburg, Germany, Federal Republic of Hoyler, Gerhard, Balanstr. 362, Munich, Germany, Federal Republic of Kessler, Hartmut, Dechbettener Str. 19, Regensburg, Germany, Federal Republic of

We claim as our invention:

plurality of planar stacked dielectric layers, a metal coating on each of the [\*1] 1. A stacked layer type capacitor for being supported on its lead wires in spaced apart openings on a printed circuit board comprising: a

LEVEL 1 - 216 OF 225 PATENTS

3,698,296

Oct. 17, 1972

ACTINIC LABEL-MAKING TOOL

INVENTOR: Heuser, Elliott G., Mequon, Wisconsin Muttera, Jr., William H., Whitefish Bay, Wisconsin

We claim:

[\*1] 1. A label-making tool adapted for the manufacture of labels from tape which has an adhesive layer and an ultraviolet-imaging layer of the type which activates to visibly change color upon exposure to ultraviolet light and deactivates upon exposure to visible light, said tool comprising, in combination:

I. a housing having a first portion adapted to ... LEVEL 1 - 217 OF 225 PATENTS

235

PAGE

3,696,499

Oct. 10, 1972

METHOD FOR MAKING A COMPOSITE TUBE

INVENTOR: Dromsky, John A., North Attleboro, Massachusetts

laminate material together, [9\*] ... [\*7] 7. A method for making a double-walled tubing comprising the steps of heating a strip of composite metal laminate material embodying a thin inner layer of Type 304 austenitic stainless steel which is sandwiched between and metallurgically bonded to two relatively thicker outer layers of Type 1008 aluminum-killed low carbon steel to a temperature in the range from about 1850oF. to about 2,005oF. for a period of time in the range from about one-half

LEVEL 1 - 218 OF 225 PATENTS

3,688,395

Sep. 5, 1972

CONSTRUCTION METHOD OF MAKING ELECTRICAL CONNECTION

INVENTOR: Cummings, Harold K., Whitewater, Wisconsin

... [\*1] contact therewith.

2. The invention in accordance with claim 1,

wherein said base member has an outer conductive surface on which said cutting means is formed, and wherein said insulated wire is wound to form a coil of the multi-layer type.

3. The invention in accordance with claim 1, \_\*3

wherein said base member is a terminal to which said insulated wire is to be electrically connected.

4. A method of making a multi-layer electrical ... [\*4]

37

LEVEL 1 - 219 OF 225 PATENTS

3,664,973

May 23, 1972

HYDROTHERMAL METHOD FOR MANUFACTURING A NOVEL CATALYTIC MATERIAL, CATALYSTS CONTAINING SAID MATERIAL, AND PROCESSES USING SAID CATALYSTS

INVENTOR: Jaffe, Joseph, Berkeley, California

What is claimed is:

[\*1] 1. A synthetic layer-type, crystalline, clay-like mineral having the empirical formula:

nSi02 : Al203 : mAB : xH20

where the layer lattices comprise said silica, said alumina, and said B, and where

n is from 0.4 to 15.0

m is from 0.2 to 0.6

... [\*7] hydrogenating component precursor selected from compounds of Group VI metals and compounds of Group VIII metals.

[\*8] 8. A hydrocarbon conversion catalyst cracking component material obtained by the dehydration of a synthetic layer-type, crystalline, clay-like mineral having the empirical formula:

nSi02 : Al203 : mAB : xH20

where the layer lattices comprise said silica, said alumina, and said B, and where

n is from 0.4 to 15.0

m is from 0.2 to 0.6

... [\*12] a hydrogenating component precursor selected from compounds of Group VI metals and compounds of Group VIII metals.

[\*13] 13. A process of preparing a catalytic component material which comprises dehydrating a synthetic layer-type, clay-like, crystalline mineral having the empirical formula:

nSi02 : AL203 : mAB : xH20

238 where the layer lattices comprise said silica, said alumina, and said B, and

Pat. No. 3664973, \*13

n is from 0.4 to 15.0

m is from 0.2 to 0.6

PAGE

LEVEL 1 - 220 OF 225 PATENTS

3,626,352

Dec. 7, 1971

ATTENUATOR SWITCHES HAVING DEPOSITED LAYER-TYPE CIRCUITRY

INVENTOR: McCoig, Kenneth W., Anaheim, California

.. [\*2] said second wafer being movable relative to said first wafer,

attenuation circuit comprising a resistor array having deposited-layer-type an attenuation circuit disposed on a face of said first wafer, said resistors and conductors, contact means, attached to said first wafer, for making electrical connection to conductors of said resistor array, said contact means having contact ends spring biased against said ...

- $\ldots$  [\*2] circuit depending on the relative orientation of said first and second wafers.
- $[\star 3]$  3. An attenuator switch as defined in claim 2 wherein said electrically insulative material comprises a ceramic and wherein said deposited layer-type resistors are fabricated of cermet or conductive plastic.
- [\*4] 4. An attenuator switch as defined in claim 2 wherein said resistor array comprises first, second, and third resistors connected as a pi ...
- spaced parallel relationship with a plurality of rotary wafers, ... [\*11]

a shaft extending through an opening in the middle of each stationary wafer and cooperating to rotate simultaneously all of said rotary wafers,

a deposited layer-type attenuator section disposed on each of said stationary wafers, each attenuator section comprising deposited layer-type resistors and conductors.

a set of deposited layer-type conductive switch pads disposed on each rotary wafer, and

spring-metal contacts extending from each stationary wafer and electrically connected to the attenuator section thereof, said contacts cooperating with switch pads on an associated rotary wafer to insert or bypass said section depending on the rotational position of said shaft.

- $[*12] \quad 12. \ A$  step attenuator switch as defined in claim 11 wherein said deposited layer-type attenuator section is disposed on the front face of said stationary wafer and wherein ends of said contacts project rearwardly of said wafer through spaced holes therein.
- PAGE [\*13] 13. A step attenuator switch as defined in claim 11 wherein said deposited layer-type attenuator section is disposed on the front face of said stationary wafer and wherein ends of said contacts project forwardly of said

Pat. No. 3626352, \*13

- [\*14] 14. A step attenuator switch as defined in claim 11 wherein ...
- ... [\*14] each contact being attached by a fastener spaced a selected distance from a free end of said each contact,

said attenuator section comprising first, second, and third deposited layer-type resistors connected in pi configuration, a first deposited layer-type conductor connecting the junction of said first and second resistors to one of said contacts, a second deposited layer-type conductor connecting the junction of said second and third resistors to a second of said contacts, the junction of said first and third resistors being connected to a common terminal by a third deposited layer-type conductor, and

a pair of input/output terminals electrically connected respectively to said third and said fourth contacts.

- [\*15] 15. A bridged-T attenuator comprising:
- a wafer of electrically insulative material,

portions of a bridged-T circuit disposed on both major faces of said wafer and formed of deposited layer-type components, and wiper contact means rotatable with respect to said wafer and cooperating with said deposited layer-type components for controlling the attenuation of said

16. A bridged-T attenuator as defined in claim 15 wherein said wafer comprises a refractory material and has a central opening ... attenuator further comprising an input terminal, an output terminal and a common terminal all attached to said wafer, a pair of deposited layer-type fixed resistors being connected in series by means of deposited layer-type conductors between said input and output terminals. 17. A bridged-T attenuator as defined in claim 16 further comprising [\*17]

major face of said wafer surrounding said opening and electrically connected by means of a deposited layer-type conductive strip to said input terminal, a first annular deposited layer-type conductive switch pad disposed on one

a first plurality of deposited layer-type conductive switch pads disposed in a circle on said one major face surrounding said central opening, a pair of said first plurality of switch pads being electrically connected respectively to said input and output terminals, and

a first plurality of deposited layer-type relative elements disposed on said one major face and electrically connected between adjacent ones of said first plurality of switch pads, said wiper contact means selectively electrically shorting one of ...

... [\*17] controlling the effective resistance of one portion of said bridged-T circuit.

Pat. No. 3626352, \*17

18. A bridged-T attenuator as defined in claim 17 further comprising:

a second annular deposited layer-type conductive switch pad disposed on the other major face of said wafer surrounding said opening and electrically connected by means of a deposited layer-type conductive strip to the junction of said pair of fixed resistors, a second plurality of deposited layer-type conductive switch pads disposed in a circle on said other major face surrounding said central opening

a second plurality of deposited layer-type resistive elements disposed on said other major face, one of said second plurality of resistive elements being electrically connected between one of said second plurality of switch pads and

LEVEL 1 - 221 OF 225 PATENTS

3,617,491

Nov. 2, 1971

HYDROCRACKING CATALYST COMPRISING A LAYERED CLAY-TYPE CRYSTALLINE ALUMINOSILICATE COMPONENT, A GROUP VIII

PAGE 241

AGE 242

## COMPONENT AND A THORIUM OR URANIUM COMPONENT, AND PROCESS USING SAID CATALYST

INVENTOR: Csicsery, Sigmund M., Lafayette, California

 $\dots$  [\*5] metals, and wherein said hydrogenating components are contained in said matrix.

6. A catalyst as in claim 5, which further comprises Titania. [46]

[\*7] 7. A catalyst comprising:

A. A dehydrated layer-type, crystalline, claylike mineral-cracking component which prior to dehydration has the empirical formula nSi02 :A1 203 :mAB: xH20,

where the layer lattices comprise said silica, said alumina, and said B, and

n is from 2.4 to ...

LEVEL 1 - 222 OF 225 PATENTS

243

3,617,490

Nov. 2, 1971

HYDROCRACKING CATALYST COMPRISING A LAYERED CLAY-TYPE CRYSTALLINE ALUMINOSILICATE COMPONENT, A GROUP VIII COMPONENT, AND A CHROMIUM OR TUNGSTEN COMPONENT, AND PROCESS USING SAID CATALYST

INVENTOR: Csicsery, Sigmund M., Lafayette, California

... [ $\star 5$ ] metals, and wherein said hydrogenating components are contained in said matrix.

[\*6] 6. A catalyst as in claim 5, which further comprises titania.

A catalyst comprising: [\*7] A. A dehydrated layer-type, crystalline, claylike mineral cracking component which prior to dehydration has the empirical formula nSi02:Al203:mAB:xH20,

where the layer lattices comprise said silica, said alumina, and said B, and

n is from 2.4 to 3.0

244

LEVEL 1 - 223 OF 225 PATENTS

PAGE

3,617,489

Nov. 2, 1971

HYDROCRACKING CATALYST COMPRISING A LAYERED CLAY-TYPE CRYSTALLINE ALUMINOSILICATE COMPONENT, A GROUP VIII. COMPONENT AND GOLD, PROCESS USING SAID CATALYST

INVENTOR: Csicsery, Sigmund M., Lafayette, California

... [\*6] metals, and wherein said hydrogenating components are contained in said matrix.

[\*7] 7. A catalyst as in claim 6, which further comprises titania.

8. A catalyst comprising:  A. a dehydrated layer-type, crystalline, claylike mineral cracking component which prior to dehydration has the empirical formula

nSi02 : A1203 : mAB : xH20,

where the layer lattices comprise said silica, said alumina, and said B, and

n is from 2.4 to 3.0

245

PAGE

3,615,501

LEVEL 1 - 224 OF 225 PATENTS

Oct. 26, 1971

COLOR PHOTOGRAPHIC DEVELOPING PROCESS

INVENTOR: Ohi, Reiichi, Kanagawa, Japan

Shimamura, Isao, Kanagawa, Japan Shishido, Tadao, Kanagawa, Japan

- ... [\*4] black and white developer or in a pretreatment bath before the black and white development.
- [\*5] 5. The process according to claim 1 wherein said multilayer color photographic element is a coupler-in-emulsion layer-type color photographic element and said compound is incorporated in the black and white developer.
- [\*6] 6. The process according to claim 1 wherein said process is a high temperature process conducted at a temperature higher than ...

LEVEL 1 - 225 OF 225 PATENTS

3,611,078

Oct. 5, 1971

### STABILIZED AC SUPERCONDUCTOR

INVENTOR: Massar, Ernst, Erlangen, Germany, Federal Republic of Parsch, Claus-Peter, Erlangen, Germany, Federal Republic of

We claim:

- [\*1] 1. An AC superconductor, comprised of a superconducting layer of type I intended for the load current, which is placed with a minimum contact resistance upon a metallic stabilizing layer of a superconducting material of type III, which during overloading absorbs the current, at least partially and temporarily, said superconductor of type I encloses said superconductor of type II provided for stabilizing purposes in the form of a tube.
- [\*2] 2. The superconductor of claim 1 wherein the superconducting layer of type I is lead.
- [\*3] 3. The superconductor of claim 1, wherein the superconductors are concentric tubes.
- [\*4] 4. The superconductor of claim 3, wherein at least two mutually contacting layers of superconducting material of type III, provided for stabilization, are ...
- $\cdots$  [\*7] tube upon whose outer wall the superconductor layers are placed.
- $[^{*8}]$  8. The superconductor of claim 5, wherein the thickness of the respective superconducting layer is between 1 and 10 mu .
- [\*9] 9. An AC superconductor, comprised of a superconducting layer of type II intended for the load current, which is placed with a minimum contact resistance upon a metallic stabilizing layer of a superconducting material of type III, which during overloading absorbs the current at least ...
- ... [\*11] III, provided for stabilization, are present which have higher critical field strengths for the alternating current the further they are from the superconductor of type II which is provided for the current load.
- [\*12] 12. The superconductor of claim 9 wherein the superconducting layer of type II is niobium.
  - 12:38 P.M. STARTED 246 PAGES
  - 68990

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P.O. BOX 218
YORKTOWN HEIGHTS NEW YORK 10598-0218
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CLAIMS(LAYER-TYPE OR (LAYER PRE/1 TYPE))
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5,028,786

GET 1st DRAWING SHEET OF

Jul. 2, 1991

Array for a nuclear radiation and particle detector

INVENTOR: Da Silva, Angela J., Vancouver, Canada Le Gros, Mark A., Vancouver, Canada Turrell, Brian G., Vancouver, Canada Kotlicki, Andrzej, Warsaw, Maryland, Poland Drukier, Andrzej K., Greenbelt, Maryland

... K) and a melting point of less than 1000c C., and more preferably less than 500c C.

The present invention also relates to a method of making an array comprising depositing a substantially continuous film layer of type I superconducting material on a substrate, removing a portion of said film to leave a plurality of discrete pixels each of a predetermined size of said type I superconducting material arranged ...

detector as defined in claim 8 wherein said each array is a ... [\*13] planar array. [\*14] 14. A method of making a detector array comprising depositing a substantially continuous film layer of type I superconducting material on a substrate removing a portion of said film to leave a plurality of discrete pixels each of a predetermined size of said type I superconducting material arranged ...

FOCUS - 2 OF 5 PATENTS

3,930,903

GET 1st DRAWING SHEET OF 1

Jan. 6, 1976

Stabilized superconductive wires

INVENTOR: Randall, Robert N., Wayland, Massachusetts Wong, James, Wayland, Massachusetts

What is claimed is:

[\*1] 1. Superconductive multi-filament wire product comprising,

means defining a copper matrix with a plurality of spaced filaments therein,

each of the filaments comprising a layer therein of type II superconducting intermetallic compound of Beta-Wolfram structure, being the diffusion reaction product of source filaments comprising a cross-section multilayer configuration of a refractory metal layer each containing ... FOCUS - 3 OF 5 PATENTS

3,910,862

GET 1st DRAWING SHEET OF

Oct. 7, 1975

Stabilized superconductors

INVENTOR: Wong, James, Wayland, Massachusetts

What is claimed is:

[\*1] 1. Superconductive multi-filament wire product comprising,

means defining a bronze matrix with a plurality of spaced filaments therein,

each of the filaments comprising a layer of type II superconducting intermetallic compound of Beta-Wolfram structure being the diffusion reaction product of a first elemental component derived from said bronze matrix and of second elemental component derived from source ... FOCUS - 4 OF 5 PATENTS

3,720,847

m GET 1st DRAWING SHEET OF

Mar. 13, 1973

POWER CURRENT CRYOTRON WITH FLAT GATE CONDUCTOR

INVENTOR: Massar, Ernst, Erlangen, Germany, Federal Republic of

... member. The tubular insulating member and the superconducting layer are of meander configuration so that during operation of the cryotron adjacent portions of the layer conduct current in opposite directions. This provides a power current cryotron with a layer type gate conductor superconducting layer having a thickness in the order of magnitude of the depth of penetration of a magnetic field into the superconducting layer.

The insulating member may comprise insulating material of cylindrical configuration or a ...

I claim:

PAGE

- layer type gate conductor superconducting layer on the insulating member, said layer having a thickness in the order of magnitude of the depth of penetration of a magnetic field into the superconducting layer, said insulating member and 1. A power current cryotron comprising an insulating member and a said layer ...
- during operation of said cryotron adjacent portions of said layer ... [\*1] during operation of said conduct current in opposite directions.
- [\*2] 2. A power current cryotron comprising a tubular insulating member having an axis and a layer type gate conductor superconducting layer on said insulating member, said layer having a thickness in the order of magnitude of the depth of penetration of a magnetic field into the superconducting layer, said insulating member and said ...

3,611,078

Oct. 5, 1971

### STABILIZED AC SUPERCONDUCTOR

INVENTOR: Massar, Ernst, Erlangen, Germany, Federal Republic of Parsch, Claus-Peter, Erlangen, Germany, Federal Republic of

Described is an AC superconductor, comprised of a superconducting layer of type I or II intended for the load current, which is placed with a minimum contact resistance upon a metallic stabilizing layer which during overloading absorbs the current at least partially and temporarily. The stabilizing layer is

SUM:

Our invention relates to an AC (alternating current) superconductor, comprised of a superconducting layer of type I or II, provided for a charge current. This layer is applied with a minimum contact resistance upon a metallic stabilizing layer which, during certain periods, absorbs, at least partially, the current ...

DETDESC:

... stabilizing layer, comprised of superconducting material of type III, with a higher critical field intensity for the alternating current. The last-mentioned layer is permitted to have considerably higher losses during an AC load than the layers of type I or II.

Suitable stabilizing layers are, fundamentally, all conventional type III superconductors, e.g. technetium. Niobium/zirconium, for instance, has a critical field strength of 1500 to 2000 Oe for alternating current of ... ... especially preferred, since they can be cooled from the inside and from the outside, for example by means of liquid helium.

2

According to another development of the invention, especially in the case of tubular AC superconductors, the outer layer of type I or II as well as the successive inner layer of type III need not be thicker, with respect to current carrying capacity, than a few mu, e.g. 1 to 10 mu. Preferably, both layers, e.g. niobium and technetium, are placed upon a copper or aluminum ...

is not limited to a circular, though the superconductor of the present invention outer layer 1 in the FIG. symbolizes a superconductor layer of type I or II, for example pure lead or pure niobium. Layer 2 should be a superconductor of type III, e.g. technetium, niobium-titanium, niobium-zirconium or niobium-tin. In the

precipitated out of pure niobium chloride, upon the niobium-tin layer. The stabilizing layer(s) according to the invention and the superimposed superconducting layer of type I or II can also be placed upon a carrier, by PAGE 6 ... in another coating chamber of the same furnace, niobium can be

Pat. No. 3611078, \*

FOCUS

using a plasma jet method. Electrolysis processes are also suitable for producing the superconductors, in accordance with the present invention. Technetium, ...

We claim:

- I intended for the load current, which is placed with a minimum contact resistance upon a metallic stabilizing layer of a superconducting material of type III, which during overloading absorbs the current, at least partially and temporarily, said superconductor of type I encloses said superconductor of type III provided for stabilizing purposes in the form of a tube. 1. An AC superconductor, comprised of a superconducting layer of type [\*1]
  - [\*2] 2. The superconductor of claim 1 wherein the superconducting layer of type I is lead.
- 3. The superconductor of claim 1, wherein the superconductors are concentric tubes.
- [\*4] 4. The superconductor of claim 3, wherein at least two mutually contacting layers of superconducting material of type III, provided for stabilization, are ...
- ... [\*7] tube upon whose outer wall the superconductor layers are placed.
- 8. The superconductor of claim 5, wherein the thickness of the respective superconducting layer is between 1 and 10 mu
- $[\star 9]$  9. An AC superconductor, comprised of a superconducting layer of type II intended for the load current, which is placed with a minimum contact

resistance upon a metallic stabilizing layer of a superconducting material of type III, which during overloading absorbs the current at least ...

- $\dots$  [\*11] III, provided for stabilization, are present which have higher critical field strengths for the alternating current the further they are from the superconductor of type II which is provided for the current load.
- [\*12] 12. The superconductor of claim 9 wherein the superconducting layer of type II is niobium.

  4 6 PAGES

  \* 12:46 P.M. STARTED 12:46 P.M. ENDED

### $\S^*$ ATTACHMENT B



### Jim Leonard 05/24/98 02:38 PM

To:

Daniel P Morris/Watson/IBM@IBMUS

cc:

From:

Subject: Layered Like or Type

Dan,

For Layered Like or Type, here are some article abstracts. One book was found for Layered Type.

Article listing are from a search of INSPEC on DIALOG.

If citation information is needed, let me know.

All the best,

Jim

James W. Leonard, Reference Librarian, Watson Library Services. Room 16-240 IBM TJ Watson Research Center,

Route 134, Yorktown Hts. NY 10598.

jwl@us.ibm.com

Voice=(914) 945 3468; Fax=(914) 945 4144

\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*

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### Layered like

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2642109 PY=1969 : PY=1985

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14/7/1

DIALOG(R) File 2: INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

02401641 INSPEC Abstract Number: A85032877

Title: Polymorphism of diphthalocyanine-neodymium. Molecular and crystal structure of beta phase

Author(s): Darovskikh, A.N.; Tsytsenko, A.K.; Frank-Kamenetskaya, O.V.;

Fundamenskii, V.S.; Moskalev, P.N.

Author Affiliation: Inst. of Nuçl. Phys., Acad. of Sci., Leningrad, USSR Journal: Kristallografiya vol: 29, no.3 p.455-61 Publication Date: May-June 1984 Country of Publication: USSR CODEN: KRISAJ ISSN: 0023-4761 Translated in: Soviet Physics - Crystallography vol.29, no.3 p.273-6 Publication Date: May-June 1984 Country of Publication: USA ISSN: 0038-5638 CODEN: SPHCA6 U.S. Copyright Clearance Center Code: 0038-5638/84/030273-04\$03.90 Document Type: Journal Paper (JP) Language: English Treatment: Experimental (X) Abstract: X-ray structural analysis reveals that diphthalocyanine-neodymi um, with the composition PcNdPc/sub ox/ (Pc=(C/sub 32/H/sub 16/N/sub  $8/)/\sup 2-/$ , Pc/sub ox/=(C/sub 32/H/sub 16/N/sub 8/)/sup 1-/) exists in three polymorphic modifications tetragonal alpha , orthorhombic gamma , and monoclinic beta . Determination of the crystal structure of the beta phase (P2/sub 1/ automatic diffractometer, theta -2 theta method, Mo K alpha , R=0.052) revealed that it is of the structural type Pc/sub 2/U. The sandwich molecules are packed in layers parallel to the ac plane. The metal-ligand distance in the structure of Pc/sub 2/M (where M is a metal ion) is explained by the ratio between the ionic radii (r/sub Nd/>r/sub u/>r/sub Sn/). The angle of relative rotation of the ligands is apparently determined by the character of the packing. Comparing the identity periods T/sub perpendicular to / perpendicular to the layers of molecules in the alpha , beta , and gamma modifications of diphthalocyanine-neodymium (2T/sup alpha //sub (001)/=T/sup beta //sub (001)/sin beta =T/sup gamma (101)/), one sees that the M-ligand distances are stable in these //sub structures. The relation between the periods T/sup beta //sub (100)/ approximately=T/sup beta //sub (010)/ approximately=1/2T/sub (110)//sup alpha / in the alpha and beta phases shows that the tetragonal structure is evidently layered like the beta phase. (10 Refs) \*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\* Layered type ? \*s layered()type 23991 LAYERED 419473 TYPE 80 LAYERED () TYPE S15 ?•s s15 and py=1969:1985 80 S15 PY=1969 : PY=1985 2642109 15 S15 AND PY=1969:1985 S16 ?•t 16/7/1-15 16/7/1 2:INSPEC DIALOG(R)File (c) 1998 Institution of Electrical Engineers. All rts. reserv. INSPEC Abstract Number: B86015292 02616964 Title: A study of the breakdown mechanism in dual-layer MOS capacitor Author(s): Domangue, E.; Hickman, T.; Pyle, R.; Rivera, R. Author Affiliation: Motorola Inc., Austin, TX, USA Title: 35th Electronic Components Conference (Cat. No. Conference

Country of Publication: USA

Conference Date: 20-22 May 1985 Conference Location: Washington, DC,

U.S. Copyright Clearance Center Code: 0569-5503/85/0000-0396\$01.00

85CH2184-0)

USA

p.396-9 Publisher: IEEE, New York, NY, USA

Conference Sponsor: IEEE; Electron. Ind. Assoc

Publication Date: 1985

Language: English Document Type: Conference Paper (PA) Treatment: Experimental (X)

The time to break down distribution of MOS capacitors Abstract: fabricated with a multilayer dielectric was studied. The dielectric was composed of 10 nm of thermal silicon dioxide, 15 nm of LPCVD silicon nitride, and 1-3 nm of SiO/sub 2/ thermally grown on the Si/sub 3/N/sub 4/ layer. The test capacitor was constructed with paralleled storage cells in a 64K dynamic memory device. Various electric fields and temperatures were used to stress the layered type of capacitors and a control group consisting of the same vehicle but having a 39 nm silicon dioxide dielectric. Stressed units were physically analyzed to isolate the failure sites. The type and location of the dielectric breakdown faults were found to be similar in both types of dielectric structure. The layered dielectric demonstrated superior reliability, however, which is attributed to lower defectivity or the spatial variation of the applied electric field within the structure. (9 Refs)

16/7/2 DIALOG(R) File 2:INSPEC (c) 1998 Institution of Electrical Engineers. All rts. reserv.

INSPEC Abstract Number: A85096207 Title: Reflectivity, joint density of states and band structure of group IVb transition-metal dichalcogenides

Author(s): Bayliss, S.C.; Liang, W.Y.

Author Affiliation: Cavendish Lab., Cambridge Univ., UK

Journal: Journal of Physics C (Solid State Physics) vol.18, no.17 p.3327-35

Publication Date: 20 June 1985 Country of Publication: UK

CODEN: JPSOAW ISSN: 0022-3719 U.S. Copyright Clearance Center Code: 0022-3719/85/173327+09\$02.25

Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Optical joint density of states (OJDOS) functions have been obtained from Kramers-Kronig analysis of reflectivity measurements for the layered-type materials TiS/sub 2/, TiSe/sub 2/, ZrS/sub 2/, ZrSe/sub 2/, HfS/sub 2/ and HfSe/sub 2/. The reflectivity measurements were made at near-normal incidence over the photon energy range 0.6-14 eV at 77K. Comparison of the OJDOS functions shows that there are many similarities in the band shapes which can be explained in terms of the amount of trigonal distortion present in the crystal lattice and the differences in binding energy of electron levels in the atoms. (9 Refs)

16/7/3 2:INSPEC DIALOG(R) File

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

INSPEC Abstract Number: A84041348, B84023254

Title: Hydriodic acid photodecomposition on layered-type transition metal dichalcogenides

Author(s): Bicelli, L.P.; Razzini, G.

Author Affiliation: Dept. of Appl. Phys. Chem., Milan Polytech., Milan, Italy

vol.20, no.4 p.393-403 Journal: Surface Technology

Country of Publication: Switzerland Publication Date: Dec. 1983

CODEN: SUTED8 ISSN: 0376-4583

U.S. Copyright Clearance Center Code: 0376-4583/83/\$3.00

Document Type: Journal Paper (JP) Language: English

Treatment: Experimental (X)

The photodecomposition of hydriodic acid on platinized Abstract: n-WSe/sub 2/ single crystals immersed in an aqueous 1 M HI solution was studied. During the photodecomposition process, hydrogen evolution only

occurred on the microscopic defects of the sample surface, whereas iodine was produced on the smooth areas where a diffuse orange-red colouring appeared. For polycrystalline specimens, however, hydrogen gas bubbles were formed over the entire surface, the rate of process being markedly slower than on single crystals. The results are discussed with the assumptions that the n-WSe/sub 2/ single crystals behave as Schottky-type photochemical diodes, that the cathodic reaction takes place on the stepped platinum-covered areas and that the anodic reaction occurs on the smooth unplatinized areas. (26 Refs)

16/7/4
DIALOG(R)File 2:INSPEC
(c) 1998 Institution of Electrical Engineers. All rts. reserv.

02085031 INSPEC Abstract Number: A83077984, B83041973 Title: Mechanistic studies of reversible layer-type electrodes Author(s): Rouxel, J.; Molinie, P.; Top, L.H. Author Affiliation: Lab. de Chimie des Solides, Nantes, France Journal: Journal of Power Sources vol.9, no.3-4 p.345-57 Publication Date: April-May 1983 Country of Publication: Switzerland CODEN: JPSODZ ISSN: 0378-7753 U.S. Copyright Clearance Center Code: 0378-7753/83/0000-0000/\$3.00 Conference Title: International Meeting on Lithium Batteries Conference Date: 27-29 April 1982 Conference Location: Rome, Italy Language: English Document Type: Conference Paper (PA); Journal Paper (JP)

Treatment: Theoretical (T)

Abstract: In layered type intercalation electrodes ions are stored reversibly during the functioning of secondary batteries. The behaviour of the system depends on geometrical and electronic factors. The geometrical factors are concerned with the localization of the ions in the host structure; they deal with average structure determinations and local ordering problems. The diffusion properties of the intercalated ions depend on the site geometry, the population of the Van Der Waals gap, the ionicity of the bonds in the host, the stoichiometry of the host, and the mechanical properties of its slabs. Electrons have to be accommodated by the host. The band structure of the host plays an important role in respect of the ability to intercalate, the phase limit, and the stability of the products. Metal-insulator transition may be induced. Other possible factors such as Jahn-Teller effects have also to be considered. (23 Refs)

16/7/5
DIALOG(R)File 2:INSPEC
(c) 1998 Institution of Electrical Engineers. All rts. reserv.

01994480 INSPEC Abstract Number: A83023215

Title: Structure of tungstic acids and amorphous and crystalline WO/sub 3/thin films

Author(s): Ramans, G.M.; Gabrusenoks, J.V.; Veispals, A.A.

Author Affiliation: Inst. of Solid State Phys., P. Stucka Univ., Riga, USSR

Journal: Physica Status Solidi A vol.74, no.1 p.K41-4

Publication Date: 16 Nov. 1982 Country of Publication: East Germany

CODEN: PSSABA ISSN: 0031-8965

Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: The authors compare the Raman spectra of a-WO/sub 3/ with spectra of crystalline WO/sub 3/.H/sub 2/0, WO/sub 3/.2H/sub 2/0 and amorphous bulk WO/sub 3/.H/sub 2/0. It is concluded from the results that the structure of a-WO/sub 3/ films consists of a layered type structure of tungsten hydrates and of a framework structure of tungsten anhydride. The band at 590 cm/sup -1/ is attributed to stretching modes of the terminal

74₽ 31:

oxygen. By dehydration of amorphous WO/sub 3/.1.74 H/sub 2/O one can get amorphous bulk samples with a structure similar to the a-WO/sub 3/ thin films. (12 Refs)

16/7/6
DIALOG(R)File 2:INSPEC
(c) 1998 Institution of Electrical Engineers. All rts. reserv.

01973788 INSPEC Abstract Number: A83008001

Title: Synthesis of new layered-type and new mixed-layered-type bismuth compounds

Author(s): Kodama, H.; Watanabe, A.

Author Affiliation: Nat. Inst. for Res. in Inorganic Materials, Ibaraki, Japan

Journal: Journal of Solid State Chemistry vol.44, no.2 p.169-73
Publication Date: Sept. 1982 Country of Publication: USA
CODEN: JSSCBI ISSN: 0022-4596

U.S. Copyright Clearance Center Code: 0022-4596/82/110169-05\$02.00/0 Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: Four new compounds, PbBi/sub 2/TiTaO/sub 8/F, PbBi/sub 2/TiNbO/sub 8/F, Bi/sub 5/Ti/sub 2/WO/sub 14/F, and Bi/sub 7/Ti/sub 5/O/sub 20/F, were prepared and identified by X-ray diffraction analysis. Two of them are new members of a family called layered bismuth compounds. The other two are new members of a family called mixed-layered bismuth compounds. Thermal properties of the new compounds were studied. Moreover, the possibility of the existence of other new members belonging to the family called mixed-layered bismuth compounds is discussed. (14 Refs)

16/7/7
DIALOG(R)File 2:INSPEC
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01891945 INSPEC Abstract Number: A82076639

Title: The phase relations in the Yb/sub 2/0/sub 3/-Fe/sub 2/0/sub 3/-MO systems in air at high temperatures (M: Co, Ni, Cu, and Zn)

Author(s): Kimizuka, N.; Takayama, E.

Author Affiliation: Nat. Inst. for Res. in Inorganic Materials, Ibaraki-ken, Japan

Journal: Journal of Solid State Chemistry vol.42, no.1 p.22-7 Publication Date: 15 March 1982 Country of Publication: USA

CODEN: JSSCBI ISSN: 0022-4596

Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: The phase relations in the Yb/sub 2/0/sub 3/-Fe/sub 2/0/sub 3/-CoO system at 1350 and 1300 degrees C, the Yb/sub 2/0/sub 3/-Fe/sub 2/0/sub 3/-NiO system at 1300 and 1200 degrees C, the Yb/sub 2/0/sub 3/-Fe/sub 2/0/sub 3/-CuO system at 1000 degrees C and the Yb/sub 2/0/sub 3/-Fe/sub 2/0/sub 3/-ZnO system at 1300 degrees C were determined in air by means of a classical quenching method. New layered-type compounds, YbFeCoO/sub 4/ (a=3.4295(5) AA, c=25.198(3) AA), YbFeCuO/sub 4/ (a=3.4808(2) AA, c=24.100(2) AA), and YbFeZnO/sub 4/ (a=3.4251(2) AA, c=25.282(2) AA), which are isomorphous with YbFe/sub 2/0/sub 4/ (space group: R3m; a=3.455(1) AA, c=25.109(2) AA), and a new compound, Yb/sub 2/Cu/sub 2/0/sub 5/, were obtained. In the Yb/sub 2/0/sub 3/-Fe/sub 2/0/sub 3/-NiO system, there are no quaternary compounds. (10 Refs)

16/7/8 DIALOG(R)File 2:INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

INSPEC Abstract Number: C82012609 Title: Office automation technology-storage and retrieval of information Author(s): Kurachi, T. Author Affiliation: Toshiba Corp., Ome-shi, Japan Journal: Journal of the Institute of Electronics and Communication gineers of Japan vol.64, no.2 p.143-9 Engineers of Japan Publication Date: Feb. 1981 Country of Publication: Japan CODEN: IECJAJ ISSN: 0373-6121 Language: Japanese Document Type: Journal Paper (JP) Treatment: Applications (A); Practical (P) Abstract: The file compositions ordered using link and direct using a page map and B tree type retrieval order are described. Layered type data models as in IBM's IMS, and the MRI System 2000, network type data models as in GE's IDS and Cineam Systems' TOTAL, relational type data model as in IBM's System R and Software AG's ADABAS and distributed type data base are also described. The types of retrieval and their call words are discussed and exemplified. Floppy disc, magnetic drum, magnetic disk, large capacity memory devices and backend systems and database machines are discussed. Micrographics and graphic information files are briefly discussed. Refs) 16/7/9 2:INSPEC (c) 1998 Institution of Electrical Engineers. All rts. reserv.

DIALOG(R)File

INSPEC Abstract Number: A80098966

Title: A method of measurement of the refractive indices of crystals with layered structure

Author(s): Allakhverdiev, K.R.; Guliev, R.I.; Salaev, E.Yu.; Kulevskii, L.A.; Savelev, A.D.; Smirnov, V.V.

Author Affiliation: Inst. of Phys., Acad. of Sci., Baku, Azerbaidzhan SSR, USSR

Journal: Physica Status Solidi A vol.60, no.1 p.309-12

Publication Date: 16 July 1980 Country of Publication: East Germany

CODEN: PSSABA ISSN: 0031-8965

Document Type: Journal Paper (JP) Language: English

Treatment: New Developments (N); Experimental (X)

Abstract: A method of determining the refractive indices of the ordinary (n/sub o/) and extraordinary (n/sub e/) rays in crystals with layered type structure are described. The refractive indices of layered CdInGaS/sub 4/ and TlInS/sub 2/ are measured using this technique with the help of laser radiation source at 0.63, 1.15, and 3.39 mu m. The experimentally obtained values of n/sub o/ and n/sub e/ are extrapolated from 0.6 to 4.0 mu m by the formulas n/sub o//sup 2/=A+B(1ambda/sup 2/+C); n/sub e//sup 2/=K+L/(1ambda/sup 2/+C)lambda /sup 2/+M). The values of the extrapolation coefficients A, B, C, K, L, and M for CdInGaS/sub 4/ and TlInS/sub 2/ crystals are obtained using the electronic computer Mir-2. (4 Refs)

16/7/10 DIALOG(R)File 2:INSPEC (c) 1998 Institution of Electrical Engineers. All rts. reserv.

01496111 INSPEC Abstract Number: B80019231 Title: Fabrication of 8 turn multi-track thin film heads Author(s): Hanazono, M.; Kawakami, K.; Narishige, S.; Asai, O.; Kaneko, E.; Okuda, K.; Ono, K.; Tsuchiya, H.; Hayakawa, W. Author Affiliation: Hitachi Res. Lab., Hitachi Ltd., Ibaraki, Japan Journal: IEEE Transactions on Magnetics vol.MAG-15, no.6 Publication Date: Nov. 1979 Country of Publication: USA CODEN: IEMGAQ ISSN: 0018-9464 Conference Title: Joint INTERMAG-MMM Conference

Conference Sponsor: IEEE

Conference Date: 17-20 July 1979 Conference Location: New York, NY, USA

Language: English Document Type: Conference Paper (PA); Journal Paper (JP)

Treatment: Practical (P)

Abstract: To obtain high bit and high track densities, fabrication of thin film magnetic recording heads have been studied by a number of companies. The authors describe a newly developed method for fabricating layered type, multi-turn, multi-track thin film inductive heads with a central tap by using photolithographic and thin film deposition techniques. (6 Refs)

16/7/11
DIALOG(R)File 2:INSPEC
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01406419 INSPEC Abstract Number: A79086309

Title: A theoretical study of the effects of various laryngeal configurations on the acoustics of phonation

Author(s): Titze, I.R.; Talkin, D.T.

Author Affiliation: Sensory Communication Res. Lab., Gallaudet Coll., Washington, DC, USA

Journal: Journal of the Acoustical Society of America vol.66, no.1 p.60-74

Publication Date: July 1979 Country of Publication: USA

CODEN: JASMAN ISSN: 0001-4966

Language: English Document Type: Journal Paper (JP)

Treatment: Theoretical (T)

Abstract: Simulation of glottal volume flow and vocal fold tissue movement was accomplished by numerical solution of a time-dependent which nonuniform, orthotropic, linear, value problem in boundary incompressible vocal fold tissue media were surrounded by irregularly shaped boundaries, which were either fixed or subject to aerodynamic stresses. Spatial nonuniformity of the tissues was of the layered type, including a mucosal layer, a ligamental layer, and muscular layers. Orthotropy was required to stabilize the vocal folds longitudinally and to accommodate large variations in muscular stress. Incompressibility and vertical motions at the glottis played an important role in producing and sustaining phonation. A nominal configuration for male fundamental speaking pitches was selected, and the regulation of fundamental frequency, intensity, average volume flow, and vocal efficiency was investigated in terms of variations around this nominal configuration. Vocal intensity and efficiency are shown to have local maxima as the configurational parameters are varied one at a time. It appears that oral acoustic power output and vocal efficiency can be maximized by proper adjustments of longitudinal tension of nonmuscular (mucosal and ligamental) tissue layers in relation to muscular layers. Quantitative verification of the 'body-cover' theory is therefore suggested, and several further implications with regard to control of the human larynx are considered. (17 Refs)

16/7/12
DIALOG(R)File 2:INSPEC
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01295844 INSPEC Abstract Number: A79010903

Title: Optical phonons in TlInS/sub 2/

Author(s): Allakhverdiev, K.R.; Adigezalov, U.V.; Nani, R.Kh.; Yusifov, Yu.G.

Journal: Izvestiya Akademii Nauk Azerbaidzhanskoi SSR, Seriya Fiziko-Tekhnicheskikh i Matematicheskikh Nauk no.1 p.21-5

Publication Date: 1978 Country of Publication: USSR

CODEN: IAFMAF ISSN: 0002-3108

Language: Russian Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: The optical phonons of a wide gap semiconducting TlIns/sub 2/which has a layered type structure have been investigated by the method of long-wavelength infra-red (JR) and Raman scattering spectroscopy. The splitting of absorption bands is observed when the crystals are cooled down to 100K. The comparison of phonon frequencies determined from JR and Raman experiments revealed TlIns/sub 2/ to be centresymmetric. (10 Refs)

16/7/13

DIALOG(R) File 2:INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

01081136 INSPEC Abstract Number: A77063130

Title: Field dependence of the susceptibility maximum for two-dimensional antiferromagnet

Author(s): Mostafa, M.F.; Semary, M.A.; Ahmed, M.A.

Author Affiliation: Dept. of Phys., Faculty of Sci., Cairo Univ., Cairo, Egypt

Journal: Physics Letters A vol.61A, no.3 p.183-4

Publication Date: 2 May 1977 Country of Publication: Netherlands

CODEN: PYLAAG ISSN: 0375-9601

Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: The magnetic susceptibility measurements on layered type structure (CH/sub 3/NH/sub 3/)/sub 2/FeCl/sub 2/Br/sub 2/ revealed a transition temperature T/sub N/(H=0) approximately=100K. The transition temperature of (CH/sub 3/NH/sub 3/)/sub 2/FeCl/sub 4/ was previously found to be T/sub N/(H=0) approximately=95K. The effect of magnetic field on the transition temperature and peak intensity for both compounds has been investigated. (7 Refs)

16/7/14

DIALOG(R)File 2:INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

00360679 INSPEC Abstract Number: A72019924

Title: Magnetic ordering in LiCr/sub 1-x/Fe/sub x/O/sub 2/

Author(s): Tauber, A.; Moller, W.M.; Banks, E.

Author Affiliation: US Army Electronics Command, Fort Monmouth, N.J., USA

Journal: Journal of Solid State Chemistry vol.4, no.1 p.138-52

Publication Date: Jan. 1972 Country of Publication: USA

CODEN: JSSCBI ISSN: 0022-4596

Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: Magnetic ordering in the LiCr/sub 1-x/Fe/sub x/O/sub 2/ system as been investigated for polycrystal and single crystal specimens characterized by optical and X-ray diffraction techniques. Part of the 2/0-Fe/sub 2/0/sub 3/-Cr/sub 2/0/sub 3/ system was also Li/sub investigated. Magnetization and susceptibility measurements from 4.2 to 900K and Mossbauer measurements from 4.2 to 300K indicate that all rocksalt (space group R3m) compositions of ordered antiferromagnetically at low temperatures. The first-order phase transition tracked with all Mossbauer parameters. The Weiss molecular field theory for layered-type antiferromagnet was fitted with two exchange constants. The dependence of theta on x was found to be theta = theta /sub  $a/(1-x)/\sup 2/+$ theta /sub b/2x(1-x)+ theta /sub c/x/sup 2/, where - theta /sub a/=Cr/sup 3+/-Cr/sup 3+/ interaction, + theta /sub b/=Fe/sup 3+/-Cr/sup 3+/ interaction and - theta /sub c/=Fe/sup 3+/-Fe/sup 3+/ interaction. A magnetization associated with iron-substituted crystals originated with an epitaxial overgrowth of LiCr/sub 4.75/Fe/sub 0.25/0/sub 16/7/15

DIALOG(R)File 2:INSPEC

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INSPEC Abstract Number: C71019443

A static and dynamic finite element shell-analysis with Title: experimental verification

Author(s): Klein, S.

Author Affiliation: Aerospace Corp., San Bernardino, CA, USA

Journal: International Journal for Numerical Methods in Engineering

vol.3, no.3 p.299-316

Publication Date: July-Sept. 1971 Country of Publication: UK

CODEN: IJNMBH ISSN: 0029-5981

Language: English Document Type: Journal Paper (JP)

Treatment: Theoretical (T)

Abstract: A system of finite element shell analysis codes, called SABOR/DRASTIC, is used to analyse a complex two-layered shell of revolution under static and dynamic asymmetric loads. The dynamic analysis is compared with experimentally measured response. In this linear elastic analysis, emphasis is placed on the inherent flexibility of the finite element method in modelling the complex structural geometry of a given test specimen. Static studies, which involve variations in important shell parameters, and dynamic studies, which provide a successful correlation with experiment, are used to illustrate both the detail and the generality with which shell analyses may now be performed with confidence.

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Layered Like books = 0

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Searching ... SEARCH RESULTS

Search ID	Records Found	Search Term
S43 S44 S45 S46	1440 57219 0	layered-like layered like (layered-like) or (layered w like)

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47=> f (layered-type) or (layered w type) Searching ..

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S48
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S50
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51 \Rightarrow f s50 and yr < 1986
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Search
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S51
                  s50 and yr < 1986
52=> d s51 1 f8
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Page: 1 of 1
AN: 23935341
AU: Lee, Harry Nai-Shee, 1942-
TI: Electrical transport properties of some hexagonal layered type
    transition metal chalcogenides.
YR:
    1969
LN:
    English
PT:
    Book
    ix, 83 1. charts, diagrs. 28 cm.
PH:
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3

# ATTACHMENT C

To:

Daniel P Morris/Watson/IBM@IBMUS

cc: From:

Subject: Rare Earth Like or Type

Dan,

For Rare Earth Like or Type, here are some article abstracts. No books were found.

Article listing are from a search of INSPEC on DIALOG.

If citation information is needed, let me know.

All the best,

```
Jim
```

James W. Leonard, Reference Librarian, Watson Library Services. Room 16-240 IBM TJ Watson Research Center,

Route 134, Yorktown Hts. NY 10598.

iwl@us.ibm.com

Voice=(914) 945 3468; Fax=(914) 945 4144

\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*

File 2:INSPEC 1969-1998/May W3

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# Rare Earth like

?•t 6/7/1-4

6/7/1

DIALOG(R) File 2: INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

01990663 INSPEC Abstract Number: A83018861

Title: Rare earths and actinides

Author(s): Cogblin, B.

Author Affiliation: Lab. de Phys. des Solides, Univ. Paris-Sud, Centre d'Orsay, Orsay, France

Journal: Journal of Magnetism and Magnetic Materials vol.29, no.1-3 p.1-19

Publication Date: Oct. 1982 Country of Publication: Netherlands CODEN: JMMMDC ISSN: 0304-8853

U.S. Copyright Clearance Center Code: 0304-8853/82/0000-0000/\$02.75

Conference Title: Proceedings of the 4th European Conference on Rare Earths and Actinides

Conference Date: 28-31 March 1982 Conference Location: Durham, UK Language: English Document Type: Conference Paper (PA); Journal Paper (JP)

Treatment: Bibliography (B); General, Review (G)

Abstract: Reviews the different properties of rare-earths and actinides, either as pure metals or as in alloys or compounds. Three different cases are considered: (i) in the case of 'normal' rare-earths which are characterized by a valence of 3, the author discusses essentially the magnetic ordering, the coexistence between superconductivity and magnetism and the properties of amorphous rare-earth systems; (ii) in the case of 'anomalous' rare-earths, 'intermediate-valence' systems and 'Kondo' systems are distinguished. Special emphasis is given to the problems of the 'Kondo lattice' (for compounds such as CeAl/sub 2/, CeAl/sub 3/ or CeB/sub 6/) or the 'Anderson lattice' (for compounds such as TmSe). The problem of neutron diffraction in these systems is also discussed; and (iii) in the case of actinides, the d-f hybridized and almost magnetic metals at the beginning of the series are separated from the rare-earth like metals after americium. (193 Refs)

6/7/2 DIALOG(R)File 2:INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

01381163 INSPEC Abstract Number: A79069701

Title: Magnetic properties of amorphous alloys of Fe and La, Lu, Y, and Zr Author(s): Heiman, N.; Kazama, N.

Author Affiliation: IBM Res. Lab., San Jose, CA, USA

Journal: Physical Review B (Condensed Matter) vol.19, no.3 p. 1623-32

Publication Date: 1 Feb. 1979 Country of Publication: USA

CODEN: PRBMDO ISSN: 0163-1829

Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: In order to study the systematics of the Fe-Fe exchange in amorphous rare-earth-Fe alloys, without the complications associated with the magnetic characteristics of the rare-earth elements, amorphous films of Fe alloyed with La, Lu, Y, and Zr have been prepared with a wide range of Fe concentrations. Magnetization and Mossbauer-effect measurements were made. The magnetic properties of the alloys depended critically on the choice of rare earth (or rare-earth-like element). YFe and LuFe alloys were found to have spin-glass characteristics while LaFe and ZrFe alloys were found to be ferromagnetic, but with evidence that exchange fluctuations were nearly as large as the average exchange. Thus the nature of the Fe-Fe exchange interaction depends critically upon the species of the rare earth. The most important parameter in determining the magnetic behavior of these alloys appears to be the size of the rare earth atom, with large rare-earth atoms resulting in a smaller ratio of exchange fluctuations to exchange. The same dependence of the magnetic properties upon rare-earth size appears to be important in the case of magnetic-rare-earth atoms; however, the effect of rare-earth-Fe exchange also becomes important and these effects are discussed. (30 Refs)

6/7/3 DIALOG(R)File 2:INSPEC (c) 1998 Institution of Electrical Engineers. All rts. reserv. 00692540 INSPEC Abstract Number: A74076053 Crystal fields in dilute rare-earth metals obtained from magnetization measurements on dilute rare-earth alloys Author(s): Touborg, P.; Hog, J. Author Affiliation: Tech. Univ., Lyngby, Denmark Journal: Physical Review Letters vol.33, no.13 p.775-8 Publication Date: 23 Sept. 1974 Country of Publication: USA CODEN: PRLTAO ISSN: 0031-9007 Language: English Document Type: Journal Paper (JP) Treatment: Experimental (X) Abstract: Measurements of the crystal field parameters of rare earth metals can be obtained by diluting the rare earths in nonmagnetic rare earth-like hosts. Alloys of terbium, dysprosium, and erbium with scandium, yttrium and lutetium hosts were prepared and crystal field parameters determined from magnetisation measurements. An unsystematic relationship was found between crystal field parameters and rare earth atomic number. ( 17 Refs) 6/7/4 DIALOG(R) File 2:INSPEC (c) 1998 Institution of Electrical Engineers. All rts. reserv. 00258862 INSPEC Abstract Number: A71036349 Title: The effective size of americium dissolved in lanthanum (and superconducting transition temperature of La-Al alloys) Author(s): Hill, H.H.; Ellinger, F.H. Journal: Journal of the Less-Common Metals vol.23, no.1 Publication Date: Jan. 1971 Country of Publication: Switzerland CODEN: JCOMAH ISSN: 0022-5088 Language: English Document Type: Journal Paper (JP) Treatment: Experimental (X) Abstract: The lattice parameters of alloys of f.c.c. beta -La containing 1.25 to 3.70 at% Am are determined by X-ray analysis. It is found that the effective ionic size of the dissolved Am is very close to that of elemental Am itself. The unusually weak depression of the superconducting transition temperature of beta -La caused by the addition of small amounts of Am is discussed. It is suggested that Am ions in La exhibit rare earth-like characteristics of trivalency and a localized d-electron configuration. ( 11 Refs) \*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\* Rare Earth type ?•s rare()earth()type 37833 RARE 103240 EARTH (January 1969) TYPE 419473 **S**7 10 RARE()EARTH()TYPE ?•s s7 and py=1969:1985 10 S7 2642109 PY=1969 : PY=1985 S8 3 S7 AND PY=1969:1985 ?•t 8/7/1-3 8/7/1

DIALOG(R) File 2: INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

INSPEC Abstract Number: A76057644

Title: Lattice parameter variations in the rare earth type B and C structures

Author(s): Ferguson, I.F.

Author Affiliation: RFL, Springfields, UK

Conference Title: UKAEA Diffraction Analysis Conference, the Role of Diffraction and Electron Analysis in the Fast Reactor

Editor(s): Ferguson, I.F.

Publisher: UKAEA, Warrington, Lancs., UK Publication Date: 1975 Country of Publication: UK

Conference Date: 15-17 Oct. 1974 Conference Location: Dounreay, UK

Document Type: Conference Paper (PA) Language: English

Treatment: Experimental (X)

The lattice parameters of monoclinic europia have been Abstract: determined and contrasted with a range of solid solutions which have the same Rare Earth type B structure as europia. These solid solutions were based upon samaria and gadolinia which lie on either side of europia in the lanthanide series. Curiously, europia does not lie where it would be expected to lie on the lattice parameter plots on the basis of its ionic radius. This is attributed to its position in the middle of the lanthanide series. For the lanthanide oxides with the cubic Rare Earth type C structure an anomaly again occurs, but this time for gadolinia. (0 Refs)

8/7/2

DIALOG(R) File 2:INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

INSPEC Abstract Number: A71048689, B71021928

Title: Cast permanent magnets of cobalt, copper, and cerium: process and performance characteristics

Author(s): Cullen, T.J.

Author Affiliation: Sel-Rex Corp., Nutley, NJ, USA

Journal: Journal of Applied Physics vol.42, no.4 p.1535-6 Publication Date: 15 March 1971 Country of Publication: USA

CODEN: JAPIAU ISSN: 0021-8979

Conference Title: 16th annual conference on magnetism and magnetic materials

Conference Date: 17-20 Nov. 1970 Conference Location: Miami Beach, FL, USA

Language: English Document Type: Conference Paper (PA); Journal Paper (JP)

Treatment: Experimental (X)

Abstract: A process for casting magnets of the cobalt, copper, rare-earth weighing as much as several pounds has been developed. The characteristics of the alloys are highly reproducible between castings. The uniformity of performance within a casting is excellent. The residual induction of a typical cerium alloy casting is 5600 G. The coercive force is 4800 Oe. The variation of induction with temperature in the region of a load line of 2, is 0.08%/ degrees C from room temperature to 100 degrees C.

8/7/3

DIALOG(R) File 2:INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

INSPEC Abstract Number: A70028540 00125174

Title: Magnetic properties of erbium ferrite

Author(s): Apostolov, A.

Author Affiliation: Sofia Univ., Bulgaria

Journal: Comptes Rendus de l'Academie Bulgare des Sciences vol.22, p.995-8 no.9

Publication Date: 1969 Country of Publication: Bulgaria

CODEN: CRABAA ISSN: 0366-8681

Language: English Document Type: Journal Paper (JP)
Abstract: Erbium ferrite ErFeO/sub 3/ is of the rare-earth type with an orthorhombic deformation and spatial group Pbnm. The authors investigate the antiferromagnetism of the rare earth around the antiferromagnetic point of Neel for erbium ions, for which they measured the magnetic properties of the substance in the entire range between 2 degrees K and 1200 degrees K.

# ATTACHMENT D

To:

Daniel P Morris/Watson/IBM@IBMUS

cc: From:

Subject: Perovskite Like and Type

Dan,

For Perovskite Like or Type, here are some article abstracts and some books.

Article listing are from a search of INSPEC on DIALOG, and the book listings are from searching the EPIC OCLC database.

1.6

If citation information is needed, let me know.

All the best,

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Jim
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James W. Leonard, Reference Librarian, Watson Library Services. Room 16-240 IBM TJ Watson Research Center, Route 134, Yorktown Hts. NY 10598. jwl@us.ibm.com

Voice=(914) 945 3468; Fax=(914) 945 4144

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File 2:INSPEC 1969-1998/May W3
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Set Items Description
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Perovskite like

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?•s perovskite()like
           8133 PEROVSKITE
         136878 LIKE
     S1
            467
                 PEROVSKITE()LIKE
?•s s1 and py=1969:1985
             467
                 S1
                 PY=1969 : PY=1985
        2642109
            127 S1 AND PY=1969:1985
?•t 2/7/1-10,117-127
2/7/1
DIALOG(R) File
                2:INSPEC
(c) 1998 Institution of Electrical Engineers. All rts. reserv.
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02785278 INSPEC Abstract Number: A87006595

Title: Crystal-chemical features and properties of layered bismuth vanadate-titanate 🤏

Author(s): Osipyan, V.G.; Savchenko, L.M.; Kostanyan, K.A.

Journal: Izvestiya Akademii Nauk SSSR, Neorganicheskie Materialy vol.21, no.11 p.1924-7

Publication Date: Nov. 1985 Country of Publication: USSR

CODEN: IVNMAW ISSN: 0002-337X

Translated in: Inorganic Materials vol.21, no.11 p.1676-9 Publication Date: Nov. 1985 Cocoden: INOMAF ISSN: 0020-1685 Country of Publication: USA

U.S. Copyright Clearance Center Code: 0020-1685/85/2111-1676\$09.50

Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: It has been established that Bi/sub 13/V/sub 5/TiO/sub 34/ belongs to the ferroelectric family of bismuth-containing compounds with a layered compound. The formula unit Bi/sub 2//1/6V5/6Ti1/6O/sub 5/2/3 corresponds to a layered structure of (Bi/sub 2/0/sub 2/)/sup 2+/(Bi1/6V5/6Ti1/6O/sub 3/2/3)/sup 2-/ with one perovskite-like layer between ions of bismuthyl (Bi/sub 2/0/sub 2/)/sup 2+/. The dielectric properties indicate that Bi/sub 13/V/sub 5/TiO/sub 34/ has ferroelectric properties. The solid-phase process of formation of the compound from a mixture of the initial oxides takes place in one stage in the temperature range 600-800 degrees C. (8 Refs)

2/7/2 DIALOG(R) File 2:INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

02777488 INSPEC Abstract Number: A87003002

Title: optical and electrophysical properties of complexly substitutes phases based on A/sub 2/B/sub 2/O/sub 7/ perovskite-like ferroelectrics

Author(s): Titov, Yu.A.; Leonov, A.P.; Sych, A.M.; Stefanovich, S.Yu.; Lashneva, V.V.; Venevtsev, Yu.N.
Author Affiliation: T.G. Shevchenko Kiev State Univ., Ukrainian SSR, USSR

Journal: Izvestiya Akademii Nauk SSSR, Neorganicheskie Materialy vol.21, no.10 p.1739-43

Publication Date: Oct. 1985 Country of Publication: USSR

CODEN: IVNMAW ISSN: 0002-337X Translated in: Inorganic Materials

vol.21, no.10 p.1515-19

Publication Date: Oct. 1985 Country of Publication: USA

ISSN: 0020-1685 CODEN: INOMAF

U.S. Copyright Clearance Center Code: 0020-1685/85/2110-1515\$09.50

Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: The purpose of the present work was to precisely determine whether the structures of the complexly substituted phases are centro- or noncentrosymmetric and to evaluate some of the ferroelectric and thermal characteristics and electrical-conduction properties. The investigations of the electrophysical characteristics were carried out on ceramic samples in the form of tablets with silver electrodes. The density of the ceramics was 0.7-0.8 of the X-ray density. The differential thermal analysis in the 1000-2400 degrees C temperature range was carried out on a system based on a high-temperature furnace with a tungsten heating element. The study of the nonlinear-optical properties of the phases synthesized was carried out by a method involving the generation of the second optical harmonic of their laser emission according to the 'reflection' scheme. Samples with a grain diameter from 10 to 20 mu were used in this case. (8 Refs)

2/7/3

DIALOG(R) File 2:INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

INSPEC Abstract Number: A86086709 02697831 Compositions and electrical properties of complex bismuth Title: layer-structured ferroelectric ceramics Author(s): Takenaka, T.; Sakata, K. Author Affiliation: Fac. of Sci. & Technol., Sci. Univ. of Tokyo, Chiba, Journal: Japanese Journal of Applied Physics, Supplement vol.24, suppl.24-3 p.117-19 Publication Date: 1985 Country of Publication: Japan CODEN: JJPYA5 ISSN: 0021-4922 Conference Title: Proceedings of the 5th Meeting on Ferroelectric Materials and their Applications (FMA-5) Conference Date: 29-31 May 1985 Conference Location: Kyoto, Japan Document Type: Conference Paper (PA); Journal Paper Language: English (JP) Treatment: Experimental (X) Abstract: The dielectric and piezoelectric properties of complex bismuth layer-structured ferroelectrics were studied. The Curie temperature T/sub c/ of solid solution systems based on PbBi/sub 2/Nb/sub 2/0/sub 9/ linearly

Abstract: The dielectric and piezoelectric properties of complex bismuth layer-structured ferroelectrics were studied. The Curie temperature T/sub c/ of solid solution systems based on PbBi/sub 2/Nb/sub 2/O/sub 9/ linearly increases as the tolerance factor for perovskite-like units of the layer structure rapidly decreases according to the increase of the substitution ion for Pb. A substitution of (NaBi)/sub 1/2/ for Pb in the solid solution Pb/sub 1-x/(NaBi)/sub x/2/Bi/sub 2/Nb/sub 2/O/sub 9/ (PNBN-100x) system gives the elevated T/sub c/ and the easy poling process: Na/sub 0.5/Bi/sub 2.5/Nb/sub 2/O/sub 9/ has a very high Curie temperature of 785 degrees C. The coupling factor k/sub 33/ of the hot-forged PNBN-50 is about 20%. (11 Refs)

2/7/4
DIALOG(R)File 2:INSPEC
(c) 1998 Institution of Electrical Engineers. All rts. reserv.

02693893 INSPEC Abstract Number: A86080922

Title: Growth and investigation of single crystals of Bi/sub 4/GeVO/sub 10.5/ and Bi/sub 8/P/sub 4-x/Ge/sub 1+x/O/sub 24-x/2/

Author(s): Firsov, A.V.; Bush, A.A.; Mirkin, A.E.; Venevtsev, Yu.N. Author Affiliation: L.Ya. Karpov Sci. Res. Physicochem Inst. Mosco

Author Affiliation: L.Ya. Karpov Sci. Res. Physicochem. Inst., Moscow, USSR

Journal: Kristallografiya vol.30, no.5 p.932-6
Publication Date: Sept.-Oct. 1985 Country of Publication: USSR

CODEN: KRISAJ ISSN: 0023-4761

Translated in: Soviet Physics - Crystallography vol.30, no.5 p.540-3 Publication Date: Sept.-Oct. 1985 Country of Publication: USA CODEN: SPHCA6 ISSN: 0038-5638

U.S. Copyright Clearance Center Code: 0038-5638/85/050540-04\$03.90 Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: By slow cooling of melts the authors have obtained single of the phases Bi/sub 4/GeVO/sub 10.5/ and Bi/sub 8/P/sub 4-x/Ge/sub 1+x/0/sub 24-x/2/ (x=0.25). They have monitored the chemical composition of the crystals, and have investigated them by X-ray diffraction, IR spectroscopy, and examination of their dielectric and pyroelectric They properties. have obtained new data crystallographic characteristics of the phases, and have found that the crystals exhibit the pyroelectric effect at room temperature. By high-temperature X-ray diffraction, in crystals of the phase Bi/sub 4/GeVO/sub 10.5/ at 550K they find a first-order phase transition between the orthorhombic and tetragonal forms. The crystals of the phase Bi/sub 4/GeVO/sub 10.5/ are ferroelectrics with a Curie point of 550K. The crystals of this phase have a similar structure to crystals of bismuth compounds with layered perovskite-like structures. (9 Refs)

2/7/5 DIALOG(R)File 2:INSPEC (c) 1998 Institution of Electrical Engineers. All rts. reserv. INSPEC Abstract Number: A86064258 Title: Thermal expansion data. VIII. Complex oxides, ABO/sub 3/, the perovskites Author(s): Taylor, D. Author Affiliation: Fairey Tecramics Ltd., Filleybrooks, Stone, UK Journal: Transactions and Journal of the British Ceramic Society vol.84, no.6 p.181-8 Publication Date: Nov.-Dec. 1985 Country of Publication: UK CODEN: TJBCAD ISSN: 0307-7357 Document Type: Journal Paper (JP) Language: English Treatment: Bibliography (B); Theoretical (T) Abstract: Gives regression data and percentage expansions for the following perovskites: AgNbO/sub 3/, AgTaO/sub 3/, BaBiO/sub 3/, BaFeO/sub 3/, BaPbO/sub 3/, BaSnO/sub 3/, BaTiO/sub 3/, BaZrO/sub 3/, BiFeO/sub 3/, CaMnO/sub 3/, Gd/sub 0.1/WO/sub 3/, KNbO/sub 3/, KTaO/sub 3/, LaAlO/sub 3/, LaCrO/sub 3/, LaMnO/sub 3/, LuCoO/sub 3/, NaMn/sub 7/O/sub 12/, MaNbO/sub 3/, Na/sub 0.8/WO/sub 3/, NdAlO/sub 3/, PbHfO/sub 3/, PbTiO/sub 3/, PbZrO/sub 3/, PrAlO/sub 3/, SrCeO/sub 3/, SrCoO/sub 3/, SrHfO/sub 3/, SrPbO/sub 3/, SrTiO/sub 3/, SrZrO/sub 3/, YCoO/sub 3/, YMnO/sub 3/, YbMnO/sub 3/, and for the perovskite-like compounds LiNbO/sub 3/ and YbMnO/sub 3/ and for the perovskite-like compounds LiNbO/sub 3/ and LiTaO/sub 3/. (119 Refs) 2/7/6 DIALOG(R)File 2:INSPEC (c) 1998 Institution of Electrical Engineers. All rts. reserv. INSPEC Abstract Number: A86058958 Title: Formation of single crystals of the perovskite-like ferroelectric Pb (Mg/sub 1/3/Nb/sub 2/3/)0/sub 3/ Author(s): Petrovskii, G.T.; Bondar', I.A.; Andreev, E.M.; Koroleva, L.N. Author Affiliation: I.V. Grebenshchikov Inst. of Silicate Chem., Acad. of Sci., USSR Journal: Izvestiya Akademii Nauk SSSR, Neorganicheskie Materialy p.1067-70 vol.20, no.6 Publication Date: June 1984 Country of Publication: USSR CODEN: IVNMAW ISSN: 0002-337X Translated in: Inorganic Materials vol.20, no.6 p.924-6 Publication Date: June 1984 Country of Publication: USA CODEN: INOMAF ISSN: 0020-1685 U.S. Copyright Clearance Center Code: 0020-1685/84/2006-0924\$08.50 Document Type: Journal Paper (JP) Language: English Treatment: Experimental (X) Single crystals of LMN were synthesized by spontaneous Abstract: crystallization from solution in a melt of lead oxide with added boron oxide. By varying the solvent content and the temperature regime over wide ranges, it was established that the optimal conditions from growing single crystals of LMN are 980-1100 degrees C and 50-60 mass % solvent. The resulting single crystals of lead magnesioniobate, when examined under the microscope, were seen to be isotropic, with good face development and high refractive index n=2.60. (7 Refs)

2/7/7
DIALOG(R)File 2:INSPEC
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02640282 INSPEC Abstract Number: A86052249
Title: Sintering and microstructure of bismuth-containing ferroelectric

ceramics Author(s): Osipyan, V.G.; Freidenfel'd, E.Zh. Author Affiliation: Riga Polytech. Inst., Latvian SSR, USSR Journal: Izvestiya Akademii Nauk SSSR, Neorganicheskie Materialy p.1211-13 Publication Date: July 1984 - Country of Publication: USSR CODEN: IVNMAW ISSN: 0002-337X Translated in: Inorganic Materials vol.20, no.7 p.1043-5 Publication Date: July 1984 Country of Publication: USA ISSN: 0020-1685 CODEN: INOMAF U.S. Copyright Clearance Center Code: 0020-1685/84/2007-1043\$08.50 Language: English Document Type: Journal Paper (JP) Treatment: Experimental (X) Abstract: In the study of the sintering kinetics for ceramics based on bismuth-containing ferroelectric compounds with a layer perovskite-like structure, th liquid phase character of the process has been established. By modification and addition of the original oxides over stoichiometry, a substantial reduction of the sintering temperature and expansion of the sintering temperature interval has been achieved for ceramics of compositions Bi/sub 3/TiNbO/sub 9/ and Na/sub 0.5/Bi/sub 4.5/Ti/sub 4/O/sub 15/. This is explained from the viewpoint of vacancy defect theory. The microstructure of bismuth-containing ferroelectric ceramics is formed from needle-shaped grains; this is due to preferential crystal growth is bismuth-containing compounds along the longer axes of the unit cells in these compounds. (8 Refs) 2/7/8 2:INSPEC DIALOG(R)File (c) 1998 Institution of Electrical Engineers. All rts. reserv. INSPEC Abstract Number: A86039596 02624075 Title: Role of steric factors in ionic mobility processes in compounds having a perovskite-like structure Author(s): Voronov, V.N.; Aleksandrov, K.S. Author Affiliation: L.V. Kirenski Inst. of Phys., Acad. of Sci., Krasnoyarsk, USSR Journal: Fizika Tverdogo Tela vol.27, no.7 p.1968-76 Country of Publication: USSR Publication Date: July 1985 CODEN: FTVTAC ISSN: 0367-3294 Translated in: Soviet Physics - Solid State vol.27, no.7 p.1182-7 Publication Date: July 1985 Country of Publication: USA ISSN: 0038-5654 CODEN: SPSSA7 U.S. Copyright Clearance Center Code: 0038-5654/85/071182-06\$03.90 Document Type: Journal Paper (JP) Language: English Treatment: Theoretical (T); Experimental (X) Abstract: In the search for new solid electrolytes, an attempt was made at a quantitative allowance for known influences on the mobility. The parameter rho /sub ij/=R/sub j/ /sup n/ Sigma /sub l=1/ /sup 3/ square root alpha /sub 1j//nR/sub i/ /sup 3/ square root alpha /sub i/ was used, representing intermediate positions of type j for a mobile ion of type i. An analysis of rho /sub ij/ in the case of halides with a perovskite-like structure showed that fluorine anions are much more mobile than cations or the other halogen anions. Ranges of existence were found for the fluorides promising as solid electrolytes, and such compounds were synthesized and investigated. In the elpasolite and cryolite structures, rho /sub ij/ has two values corresponding to the two observed types of motion. A comparison of the temperatures T/sub ij/ at which motion begins with the calculated rho /sub ij/ gave an empirical relation T/sub ij/=T/sub mp/- beta rho /sub

ij/. No dependence of beta approximately=700 degrees on the kinds of ion circumscribing the intermediate position, or on the type of the mobile ion,

was found for any of the compounds studied. (32 Refs)

2/7/9
DIALOG(R)File 2:INSPEC
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02611546 INSPEC Abstract Number: A86033506
Title: Microdomains in the reduction of Ca/sub 2/LaFe/sub 3/0/sub 8+z/Author(s): Gonzalez-Calbet, J.M.; Vallet-Regi, M.; Alario-Franco, M.A.

Journal: Journal of Solid State Chemistry vol.60, no.3 p.320-31
Publication Date: Dec. 1985 Country of Publication: USA

Author Affiliation: Dept. de Quimica Inorg., Univ. Complutense, Madrid,

CODEN: JSSCBI ISSN: 0022-4596

U.S. Copyright Clearance Center Code: 0022-4596/85\$3.00 Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: The reduction of Ca/sub 2/LaFe/sub 3/0/sub 8+z/ in the electron microscope shows this solid to decompose into Ca/sub 2/Fe/sub 2/0/sub 5/ and LaFe0/sub 3/, two perovskite-related line-phases which, under these conditions, appear to be thermodynamically more stable. In kinetic terms, the decomposition appears to be of the nucleation and growth type. Microdomains appear to be an essential characteristic of the system since they are present in both the reactants and the reaction products. Up to nine sets of structurally-related microdomains can simultaneously be present within the same crystal. This leads to quite elaborate electron diffraction patterns which can be interpreted in terms of perovskite superstructures. These results are discussed in terms of diffusion data on perovskite-like ferrites. (16 Refs)

2/7/10
DIALOG(R)File 2:INSPEC
(c) 1998 Institution of Electrical Engineers. All rts. reserv.

02600371 INSPEC Abstract Number: A86027018

Title: Crystal-chemical conditions for formation of new layered compounds of bismuth

Author(s): Korzunova, L.V.; Osipyan, V.G.; Shebanov, L.A.; Freidenfel'd, E.Zh.

Journal: Izvestiya Akademii Nauk SSSR, Neorganicheskie Materialy vol.20, no.12 p.2074-6

Publication Date: Dec. 1984 Country of Publication: USSR

CODEN: IVNMAW ISSN: 0002-337X

Translated in: Inorganic Materials vol.20, no.12 p.1813-15 Publication Date: Dec. 1984 Country of Publication: USA

Publication Date: Dec. 1984 Co CODEN: INOMAF ISSN: 0020-1685

U.S. Copyright Clearance Center Code: 0020-1685/84/2012-1813\$08.50

Language: English Document Type: Journal Paper (JP)
Treatment: Theoretical (T)

Abstract: The family of layered perovskite-like bismuth compounds (LPBC), first discussed by Aurivillius (1949), has the general formula (Bi/sub 2/0/sub 2/)/sup 2+/(A/sub n-1/B/sub n/0/sub 3n+1/)/sup 2-/ where A is Ca/sup 2+/, Ba/sup 2+/, Pb/sup 2+/ and other ions of the corresponding size; B is Ti/sup 4+/, Nb/sup 5+/, Ta/sup 5+/, W/sup 6+/ and other ions capable of forming oxygen octahedra; and n=1, 2, 3, . . . represents the number of perovskite-like layers between the bismuthyl layers (Bi/sub 2/0/sub 2/)/sup 2+/. The authors discuss problems arising in the formation of new layered bismuth compounds Bi/sub 2/A/sub n-1/B/sub n/0/sub 3n+3/

of new layered bismuth compounds Bi/sub 2/A/sub n-1/B/sub n/O/sub 3n+3/from two others with different numbers of perovskite-like layers n. (11 Refs)

2/7/117

DIALOG(R) File 2: INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

00530722 INSPEC Abstract Number: A73043864

Title: The SrMnO/sub 3-x/-Mn/sub 3/0/sub 4/ system
Author(s): Negas, T.
Author Affiliation: Nat. Bur. Stand., Washington, DC, USA
Journal: Journal of Solid State Chemistry vol.7, no.1 p.85-8
Publication Date: May 1973 Country of Publication: USA
CODEN: JSSCBI ISSN: 0022-4596
Language: English Document Type: Journal Paper (JP)
Treatment: Experimental (X)

Abstract: Phase relations were determined in the SrMnO/sub 3-x/-Mn/sub 3/O/sub 4/ system at elevated temperatures in air using quenching, gravimetric, and X-ray diffraction techniques. The system contains one intermediate compound, SrMn/sub 3/O/sub 6-x/ (0<or=x<or=0.10 between 900-1200 degrees C), which decomposes to SrMnO/sub 3-x/ plus Mn/sub 3/O/sub 4/near 1215 degrees C. The existence of an oxygen deficient SrMnO/sub 3-x/having the hexagonal 4-layer structure was confirmed. Crystals of perovskite-like SrMnO/sub 3-x/(x>0.25) were grown from its primary field located in the system.

2/7/118
DIALOG(R)File 2:INSPEC
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00506189 INSPEC Abstract Number: A73027443, B73016495

Title: Mossbauer studies of some perovskite-like layer-type ferroelectrics Author(s): Sultanov, G.D.; Mirishli, F.A.; Ismailzade, I.H.

Author Affiliation: Inst. Theoretical Problems Chem. Technol., Acad. Sci., Azerbaijan SSR, USSR

Journal: Acta Crystallographica, Section A (Crystal Physics, Diffraction, Theoretical and General Crystallography) vol.A28, pt.4, suppl. p.S241 Publication Date: 15 July 1972 Country of Publication: Denmark

CODEN: ACACBN ISSN: 0567-7394

Conference Title: 9th International Congress of Crystallography of the International Union of Crystallography. Abstracts only

Conference Sponsor: Internat. Union of Crystallography

Conference Date: 26 Aug.-7 Sept. 1972 Conference Location: Kyoto, Japan

Language: English Document Type: Conference Paper (PA); Journal Paper (JP)

Treatment: Experimental (X)

Abstract: In the layer-type ferroelectrics the entrance of the large cations A, having noticeably different from Bi/sup 3+/ polarizability and sizes, into the octahedral emptiness of the perovskite-like packages stipulates changing of the electric field gradient on the nuclei in the oxygen octahedra. This effect has been investigated by the Mossbauer spectra of Fe/sup 57/ in the layer-type ferroelectrics Bi/sub 2/Bi/sub 4/Ti/sub 3/Fe/sub 2/O/sub 18/ (A), (PrBi)Bi/sub 4/Ti/sub 3/Fe/sub 2/O/sub 18/ (B), Pr/sub 2/Bi/sub 4/Ti/sub 3/Fe/sub 2/O/sub 18/ (C) at temperatures between 80 degrees K and 1150 degrees K. For the crystals B and C the weak lines of the magnetic splitting conditioned by some second (Magnetoordered) phase are observed. According to temperature of disappearance of the quadrupole splitting in A, B, and C crystals the Curie temperatures of these ferroelectrics 1075 degrees K, 1050 degrees K and 900 degrees K have been determined. The Mossbauer measurements show some anomalous variation of the isomer shift around the Curie points.

2/7/119
DIALOG(R)File 2:INSPEC
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00504369 INSPEC Abstract Number: A73027007

Title: High pressure synthesis and crystal structure of NaMn/sub 7/0/sub 12/

Author(s): Marezio, M.; Dernier, P.D.; Chenavas, J.; Joubert, J.C.

Author Affiliation: Bell Telephone Labs., Murray Hill, NJ, USA

Journal: Journal of Solid State Chemistry vol.6, no.1 Publication Date: Jan. 1973 Country of Publication: USA

CODEN: JSSCBI ISSN: 0022-4596

Document Type: Journal Paper (JP) Language: English

Treatment: Experimental (X)

Abstract: A new compound, NaMn/sub 7/0/sub 12/ with the perovskite-like arrangement has been synthesized at 80 kbar and 1000 degrees C. This compound is cubic, a=7.3036 AA space group Im3 with four formula weights per unit cell. The structure has been solved by Patterson and Fourier synthesis and refined by least-squares based on 142 reflections. The final R and wR factors were 0.025 and 0.033, respectively. The A sites of the perovskite structure are occupied by sodium and manganese atoms in an ordered fashion. The sodium atoms are each surrounded by a 12-oxygen polyhedron whereas the manganese atoms have four nearest oxygens at 1.909 AA forming a square and four more at 2.688 AA forming a rectangle perpendicular to the square. The distortion of the oxygen network from the ideal perovskite structure is similar to that found for In(OH)/sub 3/ and Sc(OH)/sub 3/. (13 Refs)

2/7/120

DIALOG(R)File 2:INSPEC

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INSPEC Abstract Number: A72084489

Title: Infrared spectra of the rare earth perovskites LZO/sub 3/ (Z=A1, Cr, Fe, Co) (Crystal structure)

Author(s): Couzi, M.; Pham Van Huong

Author Affiliation: Univ. Bordeaux I, Talence, France

Journal: Journal de Chimie Physique et de Physico-Chimie Biologique vol.69, no.9 p.1339-47

Publication Date: Sept. 1972 Country of Publication: France

CODEN: JCPBAN ISSN: 0021-7689

Document Type: Journal Paper (JP) Language: French

Abstract: The infrared spectra, in the range from 800 to 40 cm/sup -1/ of perovskite-like aluminates LAlO/sub 3/ where L=La, Nd, Gd, Tb, Ho, Er, orthochromites LCrO/sub 3/ where L=La, Pr, Nd, Sm, Eu, Gd, Tb, Ho, Er, Tm, Yb, Lu and some similar compounds have been interpreted in connection with the structure of these crystals and by means of group factor analysis. Correlations have been established between the spectral evolution and the crystal distortion of these ionic compounds. (27 Refs)

2/7/121

2:INSPEC DIALOG(R)File

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INSPEC Abstract Number: A72025328

Title: Structural basis of ferroelectricity in the bismuth titanate family Author(s): Newnham, R.E.; Wolfe, R.W.; Dorrian, J.F.
Author Affiliation: Pennsylvania State Univ., University Park, PA, USA

Journal: Materials Research Bulletin vol.6, no.10 p.1029-40

Country of Publication: USA Publication Date: Oct. 1971

CODEN: MRBUAC ISSN: 0025-5408

Document Type: Journal Paper (JP) Language: English

Treatment: Experimental (X)

Abstract: More than fifty ferroelectrics belong to the Bi/sub 4/Ti/sub 3/O/sub 12/ family, and all consist of Bi/sub 2/O/sub 2/ layers interleaved perovskite-like M/sub n-1/R/sub n/O/sub 3n+1/ layers. Crystal structures of three members of the family have been refined from X-ray and neutron diffraction data, elucidating the distortions responsible for ferroelectricity. Bi/sub 2/Wo/sub 6/(n=1) is orthorhombic, space group B2cb; Bi/sub 3/TiNbO/sub 9/ (n=2) orthorhombic, A2/sub 1/am; Bi/sub 4/Ti/sub 3/O/sub 12/ (n=3) monoclinic, Pc, but very nearly orthorhombic, B2cb. Similar distortions occur in all three structures, with large rotational motions accompanying the polarization along a. Below the transition, a strong Bi-O bond is formed to the apex oxygen of the perovskite layer, tilting the octahedra and producing antiparallel shifts along b. Symmetry differences in the even- and odd-layered compounds can be explained by the type of strains produced in the perovskite layer. The octahedral cations (W, Ti, Nb) are the major contributors to the spontaneous polarization, moving about 0.4 AA toward an octahedral edge. (16 Refs)

2/7/122 DIALOG(R)File 2:INSPEC

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00359917 INSPEC Abstract Number: A72019058

Title: The crystal structures of orthorhombic SmAlO/sub 3/ and of trigonal NdAlO/sub 3/

Author(s): Marezio, M.; Dernier, P.D.; Remeika, J.P.

Author Affiliation: Bell Telephone Labs., Murray Hill, NJ, USA

Journal: Journal of Solid State Chemistry vol.4, no.1 p.11-19

Publication Date: Jan. 1972 Country of Publication: USA

CODEN: JSSCBI ISSN: 0022-4596

Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: The structures of NdAlO/sub 3/ and SmAlO/sub 3/ have been refined with precision from single crystal X-ray data. Both compounds have the perovskite-like arrangement. In the trigonal NdAlO/sub 3/ (space group R3c) the neodymium atoms have coordination number 12, the average Nd-O distance being 2.660 AA. The aluminum atoms are surrounded by a trigonally distorted octahedron, with an average Al-O distance of 1.896 AA. In the orthorhombic SmAlO/sub 3/, the samarium atoms are surrounded by 12-oxygen polyhedra but the coordination is slightly less than 12. The average Sm-O distance is 2.658 AA. The results incidate that with the orthorhombic to trigonal transition, the distortion of the rare earth polyhedron decreases, whereas that of the aluminium octahedron increases slightly. The overall distortion of the structure decreases. A comparison of the SmAlO/sub 3/ structure with that of its iron counterpart shows that the distortion from the ideal cubic perovskite structure is quite different. Therefore, the two compounds cannot be considered truly isostructural. (17 Refs)

2/7/123 DIALOG(R)File 2:INSPEC

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00310268 INSPEC Abstract Number: A71069671

Title: Investigation of polarization non-linearity near the phase transition in perovskite-like ferroelectrics, using an anharmonic oscillator model

Author(s): Fritsberg, V.Ya.

Book Title: Phase transitions in ferroelectrics p.7-21

Editor(s): Fritsberg, V.Ya.; Rolov, B.N.; Kruchan, Ya.Ya.

Publisher: Zinatne, Riga, Latvia, USSR

Publication Date: 1971 Country of Publication: USSR 205 pp.

Language: Russian Document Type: Book Chapter (BC)

Treatment: Theoretical (T)

Abstract: An analysis of the anharmonic oscillator model, as applied to the unit cell of perovskite-like lattices. The method of Boguslawski has been used to derive a microscopic concept of the coefficients A and B,

expanded in a series of the E=AP+BP/sup 3/ type, which is extended to the paraelectric state of the ferroelectric. The results are compared with experimental data (of Kirilov et al., Izv.AN SSSR, ser. fiz, 31, 1835, 1967) on the (Ba,Sr)TiO/sub 3/ system. (23 Refs)

2/7/124
DIALOG(R)File 2:INSPEC
(c) 1998 Institution of Electrical Engineers. All rts. reserv.

00256308 INSPEC Abstract Number: A71033281 Title: The bond lengths in LaFeO/sub 3/ Author(s): Marezio, M.; Dernier, P.D.

Author Affiliation: Bell Telephone Labs. Inc., Murray Hill, NJ, USA

Journal: Materials Research Bulletin vol.6, no.1 p.23-9
Publication Date: Jan. 1971 Country of Publication: USA

CODEN: MRBUAC ISSN: 0025-5408

Language: English Document Type: Journal Paper (JP)

Treatment: Experimental (X)

Abstract: The crystal structure of LaFeO/sub 3/ has been refined from X-ray data taken from a highly twinned crystal. The least-squares refinement was carried out on 695 independent reflections which gave an R index of 0.035. LaFeO/sub 3/ has the orthorhombic perovskite-like structure, though the rare earth polyhedron is quite distorted relative to the ideal cubic arrangement. The results indicate that in contrast to the other members of the orthoferrite series the coordination number of the lanthanum atoms is no longer 8. The Fe octahedron is only slightly distorted and the average Fe-O and O-O distances are 2.006 AA and 2.837 AA, respectively.

2/7/125 DIALOG(R)File 2:INSPEC

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00230804 INSPEC Abstract Number: A71016204

Title: High-pressure transformations in SrGeO/sub 3/, SrSiO/sub 3/, BaGeO/sub 3/, and BaSiO/sub 3/

Author(s): Shimizu, Y.; Syono, Y.; Akimoto, S.

Author Affiliation: Univ. Tokyo, Roppongi, Minato-ku, Japan

Journal: High Temperatures - High Pressures vol.2, no.1 p.113-20

Publication Date: 1970 Country of Publication: UK

CODEN: HTHPAK ISSN: 0018-1544

Language: English Document Type: Journal Paper (JP)

Abstract: The stability relations of SrGeO/sub 3/, BaGeO/sub 3/, SrSiO/sub 3/, and BaSiO/sub 3/ were studied in the range 650-1400 degrees C and 0-120 kbar. The atmospheric-pressure phases of SrGeO/sub 3/, BaGeO/sub 3/, and SrSiO/sub 3/ with the pseudowollastonite structure all transformed to a new phase with pseudo-orthorhombic symmetry at approximately 10-34 kbar. Above 50 kbar, a cubic perovskite structure of SrGeO/sub 3/ was obtained. A large density change (46% in total) was observed through the high-pressure transformations of SrGeO/sub 3/. BaGeO/sub 3/ was found to transform to 9H-type and 4H-type-like hexagonal perovskite-like structures above 95 kbar. The density increase in the transformation of BaGeO/sub 3/ from the pseudowollastonite structure to the perovskite-like structures was approximately 40%. A transformation from the atmospheric-pressure orthorhombic phase to an undetermined structure was found in BaSiO/sub 3/ at moderately high pressures. (11 Refs)

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DIALOG(R) File 2: INSPEC

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INSPEC Abstract Number: A70055719 Title: Electron diffraction investigation of phase transformations in thin films of tantalum oxide with perovskite-like structure Author(s): Khitrova, V.I.; Pinsker, Z.G. Journal: Kristallografiya p.540-6 vol.15, no.3 Publication Date: May 1970 Country of Publication: USSR CODEN: KRISAJ ISSN: 0023-4761 Language: Russian Document Type: Journal Paper (JP) Abstract: The method of electron diffraction structural analysis is applied to the investigation of phases in a series of cubic tantalum oxides with a perovskite-like structure and having a cell period of 7.75 AA. The analysis of the structure is accomplished by section of the three-dimensional Fourier potential series. It is found that because of appreciable change in the coordinates of the oxygen atoms, distortions of the structure occur, and the space group of the structure as a whole is Pmmm. Atomic coordinates are given. (11 Refs) 2/7/127 DIALOG(R)File 2:INSPEC (c) 1998 Institution of Electrical Engineers. All rts. reserv. INSPEC Abstract Number: A69035241 Title: Structure relations of hexagonal perovskite-like compounds ABX/sub 3/ at high pressure Author(s): Syono, Y.; Akimoto, S.; Kohn, K. Author Affiliation: Univ. Tokyo, Roppongi, Minato-ku, Japan Journal: Journal of the Physical Society of Japan vol.26, no.4 993-9 Publication Date: April 1969 Country of Publication: Japan CODEN: JUPSAU ISSN: 0031-9015 Document Type: Journal Paper (JP) Language: English Abstract: Phase stability relations among four hexagonal perovskite-like structures as well as the cubic perovskite structure have been studied for several oxides (BaMnO/sub 3/ and SrMnO/sub 3/) and fluorides (CsMnF/sub 3/, RbNiF/sub 3/ and TlNiF/sub 3/) at high pressure. A series of high pressure transformations are found to occur in the order of the packing sequence along the hexagonal c axis (or cubic 111 axis) of (ab), (ababcbcac), (abac), (abcacb) and (abc) with increasing pressure. This order is corresponding with the increasing order of the proportion of the cubic close-packed layers in the hexagonal close-packed structure. It is suggested that the tolerance factor of the perovskite structure and the Coulomb repulsive force play an important role in determining the crystal structure and its order in the series of phase transformations at high pressure.

#### Perovskite type

4/7/1

DIALOG(R)File 2:INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

02785243 INSPEC Abstract Number: A87007649 Title: X-ray and dielectric characteristics of new antiferroelectrics Pb(B/sub 1/2/Sb/sub 1/2/)0/sub 3/ Author(s): Danilenko, I.N.; Politova, E.D.; Abramova, A.N.; Ivanov, S.A.; Venevtsev, Yu.N. Author Affiliation: L.Ya. Karpov Sci.-Res. Phys. Chem. Inst., Moscow, USSR Journal: Izvestiya Akademii Nauk SSSR, Neorganicheskie Materialy vol.21, no.8 p.1407-10 Publication Date: Aug. 1985 Country of Publication: USSR CODEN: IVNMAW ISSN: 0002-337X Translated in: Inorganic Materials vol.21, no.8 Publication Date: Aug. 1985 Country of Publication: USA CODEN: INOMAF ISSN: 0020-1685 U.S. Copyright Clearance Center Code: 0020-1685/85/2108-1233\$09.50 Document Type: Journal Paper (JP) Language: English Treatment: New Developments (N); Experimental (X) Abstract: Compounds of the composition Pb(B/sub 1/2//sup 3+/Sb/sub 1/2/)O/sub 3/ were synthesized, where B/sup 3+/=Sc, Lu, Yb, Tm, Er, and Ho with a tendency toward the perovskite-type structure. Structural phase transformations were discovered, which are accompanied with anomalous dielectric properties. The new compounds are antiferroelectric with Curie temperatures below 330-458K. (7 Refs) 4/7/2 DIALOG(R)File 2:INSPEC (c) 1998 Institution of Electrical Engineers. All rts. reserv. 02757758 INSPEC Abstract Number: A86117797 Structural aspects of perovskite-type compounds. Symmetry changes Title: in SrCeO/sub 3/ and its solid solutions Author(s): de Pretis, A.; Minichelli, D.; Ricciardiello, F. Author Affiliation: Istituto di Chimica Applicata e Ind., Trieste Univ., Italy Journal: Revue Internationale des Hautes Temperatures et des Refractaires vol.22, no.3-4 p.215-19 Publication Date: 1985 Country of Publication: France CODEN: RIHTAV ISSN: 0035-3434 Language: English Document Type: Journal Paper (JP) Treatment: Experimental (X) Abstract: Many ABO/sub 3/ perovskite-type compounds exhibit an orthorhombic symmetry with a approximately=b and c approximately=a square root 2 axis length. An important exception is the compound SrCeO/sub 3/ which exhibits a strongly distorted perovskite-type structure with 2 a approximately=b. However, the doubling of the b axis in SrCeO/sub 3/disappears both in the SrCeO/sub 3/-SrZrO/sub 3/ and in the SrCeO/sub 3/-BaZrO/sub 3/ solid solutions in all the compositions differing from the stoichiometric SrCeO/sub 3/. (8 Refs) 4/7/3 DIALOG(R)File 2:INSPEC (c) 1998 Institution of Electrical Engineers. All rts. reserv. INSPEC Abstract Number: A86104414 02730414 Title: New seignettomagnets with perovskite type structure Yu.N.; Zhitomirsky, I.D.; Gagulin, V.V.; Author(s): Venevtsev, Sevastyanova, L.G.; Burdina, K.P. Author Affiliation: L.Ya. Karpov Inst. of Phys. Chem., Moscow, USSR Japanese Journal of Applied Physics, Supplement Journal: p.1063-5 supp1.24-2 Publication Date: 1985 Country of Publication: Japan

CODEN: JJPYA5 ISSN: 0021-4922

Conference Title: Proceedings of the Sixth International Meeting on

Ferroelectricity

Conference Sponsor: IUPAP; Int. Union Crystallogr.; Crystallogr. Soc.

Conference Date: 12-16 Aug. 1985 Conference Location: Kobe, Japan

Language: English Document Type: Conference Paper (PA); Journal Paper (JP)

Treatment: Experimental (X)

Abstract: Physical properties and phase transitions of 10 complex metal oxides of perovskite type have been studied. Some of them have been identified as seignettomagnets, ferrimagnets, antiferromagnets. (10 Refs)

4/7/4

DIALOG(R) File 2:INSPEC

(c) 1998 Institution of Electrical Engineers. All rts. reserv.

INSPEC Abstract Number: B86053876

Title: New ceramic electrode for piezoelectric buzzer

Author(s): Nomura, S.; Yoshino, H.; Yamashita, Y.

Author Affiliation: Toshiba Res. & Dev. Center, Toshiba Corp., Kawasaki,

Japanese Journal of Applied Physics, Supplement Journal: vol.24, suppl.24-2 p.736-8

Publication Date: 1985 Country of Publication: Japan

CODEN: JJPYA5 ISSN: 0021-4922

Conference Title: Proceedings of the Sixth International Meeting on Ferroelectricity

Conference Sponsor: IUPAP; Int. Union Crystallogr.; Crystallogr. Soc. Japan

Conference Date: 12-16 Aug. 1985 Conference Location: Kobe, Japan Language: English Document Type: Conference Paper (PA); Journal Paper

Treatment: Practical (P); Experimental (X)

Abstract: Electrically conductive oxide electrodes were investigated, and the piezoelectric characteristics were measured for lead zirconate titanate (PZT) resonator with the conductive oxide electrodes. Printable pastes were made using conductive oxides having perovskite type structure, such as BaPbO/sub 3/, BaPb/sub 1.2/O/sub 3/, BaPb/sub 0.8/Bi/sub 0.2/O/sub 3/, BaPb/sub 0.8/Sb/sub 0.2/O/sub 3/ and La/sub 0.5/Sr/sub 0.5/CoO/sub 3/, with glass frits and organic vehicles. the electrical resistivities of these pastes were measured at room temperature by four probe method, which were 1.4\*10/sup -2/ohm-cm for BaPbO/sub 3/ paste and 2.3\*10/sup -2/ohm-cm for La/sub 0.5/Sr/sub 0.5/CoO/sub 3/ paste. The piezoelectric resonators were obtained with Kp=39%, tan delta =3.2% and C=24nF for an electrode of BaPbO/sub 3/ paste and with Kp=31%, tan delta =6.8% and C=39nF for an electrode of La/sub 0.5/sr/sub 0.5/coo/sub 3/ paste. (8 Refs)

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DIALOG(R)File 2:INSPEC

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INSPEC Abstract Number: A86093928, B86055736 02722499

Optical detector using superconducting BaPb/sub 0.7/Bi/sub Title: 0.3/O/sub 3/ (BPB) thin films

Author(s): Enomoto, Y.; Murakami, T. Author Affiliation: Ibaraki Electr. Commun. Lab., NTT, Ibaraki, Japan Japanese Journal of Applied Physics, Supplement Journal: p.471-3 suppl.24-2

Publication Date: 1985 Country of Publication: Japan

CODEN: JJPYA5 ISSN: 0021-4922

Conference Title: Proceedings of the Sixth International Meeting on

Ferroelectricity

Conference Sponsor: IUPAP; Int. Union Crystallogr.; Crystallogr. Soc. Japan

Conference Date: 12-16 Aug. 1985 Conference Location: Kobe, Japan Document Type: Conference Paper (PA); Journal Paper Language: English (JP)

Treatment: Experimental (X)

Abstract: Highly sensitive optical detectors have been fabricated by using perovskite type superconductor BaPb/sub 0.7/Bi/sub 0.3/0/sub 3/ (BPB) thin films. Optical signals create quasi-particles and induce changes in superconducting order parameter. These changes are measured by tunneling junctions along grain boundaries in the BPB polycrystalline thin films. The sensitivity is about 10/sup 3/ V/W in the 1.0 approximately 10 mu m wavelength range and they can respond up to 600 MHz at the wavelength of 3.2 mu m. The observed results suggest that the BPB detectors are suitable for application in optical communication systems and infrared spectrometers. (7 Refs)

4/7/510 DIALOG(R) File 2:INSPEC

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00070494 INSPEC Abstract Number: A69045389

Title: Millimeter wave e.s.r. studies of ferric iron in perovskite-type oxides

Author(s): Pontin, R.G.; Slade, E.F.; Ingram, D.J.E.

Author Affiliation: Univ. Keele, UK

Journal: Journal of Physics C (Solid State Physics) vol.2, no.7 p.

Publication Date: July 1969 Country of Publication: UK

CODEN: JPSOAW ISSN: 0022-3719

Language: English Document Type: Journal Paper (JP)

Abstract: The paramagnetic resonance spectrum of Fe/sup 3+/ in lead and strontium titanates has been studied at Q band and 70GHz, and at temperatures down to 4.2 degrees k. The zero-field splittings have been measured directly giving values of 1.06+or-0.05cm/sup -1/ for lead titanate at room temperature and 2.698+or-0.006 cm/sup -1/ for strontium titanate. The spectrum for lead titanate can be fitted to a spin Hamiltonian with g/sub  $\overline{\text{mod}}$  mod /=g/sub i/=2.0, D=0.53 cm/sup -1/ and a=2.70+10/sup -2/cm/sup -1/ and a 20% increase in the zero-field splitting parameter is observed on cooling from room temperature to 77 degrees K indicating a small lattice deformation. A second transition tentatively identified as arising from a charge-compensated site having a zero-field splitting equal to 1.8+or-0.1 cm/sup -1/, has also been observed. The spectrum for strontium titanate has been studied at 35GHz and 70GHz and it is found that the Hamiltonian parameters differ slightly from those previously determined from the 10 GHz spectrum. (11 Refs)

4/7/511 DIALOG(R) File 2:INSPEC

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INSPEC Abstract Number: A69043103 00063513

Characteristic features of the dielectric polarization of Title: ferroelectric solid solutions with the perovskite-type structure at the morphotropic phase boundary and far from it

Author(s): Stolypin, Yu.E.; Isupov, V.A.

Journal: Fizika Tverdogo Tela vol.11, no.3 p.823-5 Publication Date: March 1969 Country of Publication: USSR

CODEN: FTVTAC ISSN: 0367-3294 Translated in: Soviet Physics - Solid State

Country of Publication: USA

CODEN: SPSSA7 ISSN: 0038-5654

Language: Russian Document Type: Journal Paper (JP)
Abstract: The permittivity of solid solutions (1-x)(0.6PbTiO/sub
3/+0.4PbMg/sub 0.5/W/sub 0.5/O/sub 3/)+xPbZrO/sub 3/ was determined in
static electric fields up to 20 kV/cm. It was found that the solutions near
a morphotropic (vertical) phase boundary, separating the ferroelectric
(tetragonal, x<0.30) and paraelectric (rhombohedral, x>0.30) compositions,
exhibited different field dependences of the permittivity from the
solutions far from this boundary. This was attributed to the coexistence of
mixed tetragonal and rhombohedral phases near the morphotropic boundary and
subsequent formation of boundaries between these phases.

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DIALOG(R)File 2:INSPEC
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00063406 INSPEC Abstract Number: A69042939

Title: Phase composition of reduced and reoxidized barium titanate Author(s): Arend, H.; Kihlborg, L.

Author Affiliation: Czechoslovak Academy Sci., Prague, Czechoslovakia Journal: Journal of the American Ceramic Society vol.52, no.2 p. 63-5

Publication Date: 21 Feb. 1969 Country of Publication: USA

CODEN: JACTAW ISSN: 0002-7820

Language: English Document Type: Journal Paper (JP)

Abstract: Reducing high-purity BaTiO/sub 3/ in hydrogen for 1 h at 1275 degrees C leads to an oxygen deficiency x=0.0036 in BaTiO/sub 3-x/ with maintenance of the tetragonal/cubic perovskite-type structure. Reduction at 1325 degrees C leads to x=0.0073 and brings about transformation to the hexagonal modification. Up to 1500 degrees C (x=0.0233) no further phase change occurs. Annealing the oxygen-deficient hexagonal phase in oxygen at 850 degrees C for 2 h produces stoichiometric samples which are still hexagonal, whereas the tetragonal/cubic structure is restored by heating for 1 h at 1350 degrees C. (16 Refs)

4/7/513
DIALOG(R)File 2:INSPEC
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00063265 INSPEC Abstract Number: A69042795

Title: Perovskite type structures of aluminates of rare-earth elements

Author(s): Margolis, N.V.; Udalov, Yu.P.

Journal: Kristallografiya vol.14, no.2 p.334-6

Publication Date: March 1969 Country of Publication: USSR

CODEN: KRISAJ ISSN: 0023-4761

Language: Russian Document Type: Journal Faper (JP)

Abstract: Calculations are made of the maximum relative displacement and of the force acting on the anion in rare-earth aluminates for rhombohedral and tetragonal lattice distortion. The results are plotted against the radius of the lanthanide ion; the curves are discussed, and it is concluded that rare-earth aluminates with the perovskite structure and rhombohedral lattice distortion can exist between ionic radii of 0.942 and 0.91 AA.

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DIALOG(R)File 2:INSPEC
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00060163 INSPEC Abstract Number: A69039211
Title: Crystal growth and phase transitions of CsPbCl/sub 3/Author(s): Hirotsu, S.; Sawada, S.
Author Affiliation: Tokyo Inst., Technology, Japan

Journal: Physics Letters A vol.28a, no.11 p.762-3

Publication Date: 10 March 1969 Country of Publication: Netherlands

CODEN: PYLAAG ISSN: 0375-9601

Language: English Document Type: Journal Paper (JP)

Abstract: Three phase transitions were confirmed in the perovskite-type crystal CsPbCl/sub 3/ by observing changes of conoscopic figures with temperature. Measurements of the temperature dependence of birefringence and specific heat were also performed.

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DIALOG(R) File 2: INSPEC

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00041801 INSPEC Abstract Number: A69028558

Title: Correction of dipole field due to lattice deformation of a Perovskite-type crystal

Author(s): Kinase, W.; Uemura, Y.; Kikuchi, M.

Author Affiliation: Univ., Nishiokubo, Shinjuku, Tokyo, Japan

Journal: Journal of the Physics and Chemistry of Solids vol.30, no.2 p.441-7

Publication Date: Feb. 1969 Country of Publication: UK

CODEN: JPCSAW ISSN: 0022-3697

Language: English Document Type: Journal Paper (JP)

Abstract: Correction of Lorentz field coefficients in a Perovskite-type crystal is discussed by considering orthorhombic deformation of a simple cubic lattice. If one considers the deformation of crystal structure from the simple cubic lattice to the orthorhombic lattice a change of the internal field at some respective points caused by the dipole interaction is calculated. By applying the results, quantitative discussions are possible for many phenomena accompanied by the lattice deformation namely, ferroelectricity, piezoelectricity, photoelasticity and so on. As an example calculations are made to estimate the birefringences of the BaTiO/sub 3/ and WO/sub 3/ crystals.

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Perovskite like in books

1 perovskite-like

S2 134 perovskite

S3 57219 like

1 (perovskite-like) or (perovskite w like)

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AN: 9198569

AU: Moller, Christian Knakkergard. TI: The structure of perovskite-like caesium plumbo trihalides YR: 1959 PB: Munksgaard, PL: Kobenhavn: SE: Matematisk-fysiske meddelelser / udgivet af det Kongelige Danske videnskabernes selskab ; bd. 32, nr. 2 LN: English PT: Book PH: 27 p.: ill.; 24 cm. \*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\*\* Perovskite type 6=> f (perovskite-type) or (perovskite w type) SEARCH RESULTS Search Records Search Term ID Found -------S6 25 perovskite-type S7 134 perovskite 82277 S8 type S9 30 (perovskite-type) or (perovskite w type) 10 = f s9 and yr < 1986Searching ... SEARCH RESULTS Search Records Search Term ID Found -----S10 13 s9 and yr < 198611=> d s10 1-13 f8 Record 1 of 13 Copyright 1998 OCLC AN: 31493896 AU: Galasso, Francis S. TI: Structure, properties, and preparation of perovskite-type compounds YR: 1969 PB: Pergamon Press, PL: Oxford ; New York : SE: International series of monographs in solid state physics; v. 5 LN: English
PT: Book PH: x, 207 p. : ill. Record 2 of 13 Copyright 1998 OCLC AN: 31119286 AU: Karian, Harutun G. Tight-binding energy bands of perovskite type transition metal oxides TI: YR: 1969

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oxide. YR: ? 1985 1990 SK: 89-14970 NTC English LN: Book PT: PH: 37 p. Record 8 of 13 Copyright 1998 OCLC 26413442 AU: Dougier, P. Study of the magnetic, electrical and optical properties of the TI: perovskite-type phases of strontium vanadate ( SrVO2. ? 1975 1992 YR: SK: 92-10366 NTC English LN: PT: Book PH: 16 p. Record 9 of 13 Copyright 1998 OCLC AN: 14161872 AU: Nelson, Carl W. Ferroelectricity and the chemical bond in perovskite-type oxides TI: YR: Laboratory for Insulation Research, Massachusetts Institute of PB: Technology, Cambridge, Mass. : PL: Technical report / Laboratory for Insulation Research, Massachusetts SE: Institute of Technology; 179 English LN: PT: Book 31 p.: ill.; 28 cm. PH: Record 10 of 13 Copyright 1998 OCLC AN: 10986161 Otagawa, Takaaki, 1953-AU: Electrocatalysis of oxygen evolution of perovskite-type oxides TI: YR: 1983 LN: English Book PT: xxii, 334 leaves : ill. ; 29 cm. Record 11 of 13 Copyright 1998 OCLC 6020262 AN: Michel, Christian Gabriel, 1939-AU: Structures and relationships of some Perovskite-Type compounds. TI: 1970 YR: LN: English PT: Book 89 p. PH:

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PL: [College Station, Tex.]

LN: English PT: Book

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YR: 1969

PB: Pergamon Press
PL: Oxford, New York,

SE: International series of monographs in solid state physics, v.5

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